2009 Annual Report

Center for Integrated Nanotechnologies

Center for Integrated Nanotechnologies

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From the Directors' Office

We are pleased to present this inaugural Annual Report from the Center for Integrated Nanotechnologies. The goal of this and future reports is to reach out to our entire stakeholder community, particularly our User Community, by providing this annual forum to highlight the great science in which we are engaged at CINT and the many talented scientists and support staff who make this great science possible. This has been an exciting and productive year for our Center, but also a period of transition in both personnel and operations. Several familiar faces who played seminal roles in the conception and growth of the Center have moved on to exciting new challenges within our Laboratories, and we have had the pleasure of welcoming a number of new faces to our team. Our User Program continues to be a vibrant effort capturing the most exciting elements of nanoscience, and reflects our vision and commitment to bringing an integration focus to this endeavor.

As part of our vision for nanoscience integration and in response to feedback from our User community during our recent annual workshops, we have introduced a new inititative this year, Integration Focus Activities. These efforts (p. 9) unite our scientists across our four Thrust areas with our Users to focus on larger-scale nanoscience integration challenges for specific classes of materials, systems, and phenomena. Two specific IFAs have been established and are gaining momentum, one focused on semiconductor nanowires for energy applications and another on membrane-based nanocomposites. Our goal is to use these IFAs, along with our other signature initiative, Discovery Platforms, to attract existing and new Users to engage in these larger integration challenges.

Our commitment to operational excellence and responsiveness to our User community are reflected in the changes that we enacted this year in our User program. The most notable change was a transition from a 12-month to an 18-month proposal cycle to enable our Users to make greater progress toward their research goals. We believe this will have a substantial long-term benefit to both Users and our staff.

We hope that you will find this report both informative and stimulating. We are delighted to add this to our repertoire of communication tools, and sincerely look forward sharing our research successes, news, and highlights in future years through this mechanism.

Bob Hwang
Director

David Morris
Co-Director

Tom Picraux
Chief Scientist

Neal Shinn
User Program & Outreach Manager

Left to right: David Morris, Tom Picraux, Bob Hwang, Neal Shinn

How does one…

…control energy transfer and other interactions across interfaces and over multiple length scales?

…understand and control the interactions between nanoscale building blocks to assemble specific integrated structures?

…design and exploit interactions within assembled structures to achieve new properties and particular functionalities?

These types of high-level questions serve to guide research within each of the thrust areas in CINT where specific classes of materials are investigated.

The Center for Integrated Nanotechnologies (CINT) is a Department of Energy/Office of Science Nanoscale Science Research Center (NSRC) operating as a national user facility devoted to establishing the scientific principles that govern the design, performance, and integration of nanoscale materials. Jointly operated by Los Alamos and Sandia National Laboratories, CINT explores the continuum from scientific discovery to use-inspired research, with a focus on the integration of nanoscale materials and structures to achieve new properties and performance and their incorporation into the micro- and macro worlds. Through its Core Facility at Sandia National Laboratories and its Gateway Facility at Los Alamos National Laboratory, CINT provides open access to tools and expertise needed to explore the continuum from scientific discovery to the integration of nanostructures into the micro- and macro worlds. In its overall operations, CINT strives to achieve the following goals common to all Nanoscale Science Research Centers:

1. Conduct forefront research in nanoscale science;
2. Operate as a user facility for scientific research;
3. Provide user access to the relevant BES-supported expertise and capabilities at the host national laboratory;
4. Leverage other relevant national laboratory capabilities to enhance scientific opportunities for the nanoscience user community;

These additional goals are specific to the unique CINT mission:

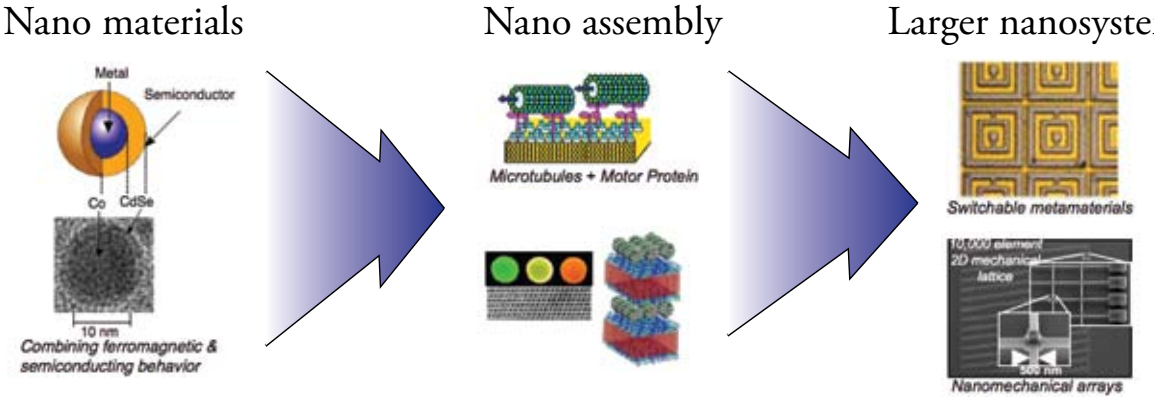
5. Establish and lead a scientific community dedicated to solving nanoscale science integration challenges;
6. Create a single user facility program that combines expertise and facilities at both Los Alamos and Sandia National Laboratories.

The CINT user program provides the international scientific community with open access to world-class scientific staff and state-of-the-art facilities for theory and simulation, nanomaterials synthesis and characterization, and unique capabilities for nanoscale materials integration, from the level of nanoscale synthesis to the fabrication of micro- and macroscale structures and devices. The staff of CINT includes laboratory scientists, postdocs and technical support staff who are leaders in the nanoscience research programs in CINT scientific thrust areas:

- Nanoscale Electronics and Mechanics,
- Nanophotonics and Optical Nanomaterials,
- Soft, Biological and Composite Nanomaterials, and
- Theory and Simulation of Nanoscale Phenomena.

The thrusts have been developed over the past several years by engaging the broader scientific community through discussions with potential CINT users and with attendees at CINT workshops, as well as by attracting some of today's top nanoscience talent to become CINT staff.

What is Nanomaterials Integration?



Advances in the development and understanding of new nanomaterials with novel behavior and response are being made at a tremendous rate. The promise of these new materials is both exciting and broad, with revolutionary implications spanning energy technologies, electronics, computing, sensing phenomena and biochemical diagnostics. However, advances in nanoscience are increasingly dependent on not just understanding the behavior of individual, isolated nanostructures, but on combining and organizing nanoscale structures to understand and discover their interactions and collective properties. Deriving the ultimate benefit from nanoscience will require the assembly of diverse nanoscale materials across multiple length scales to design and achieve new properties and functionality, in other words, nanomaterials integration.

Integration has played a pivotal and revolutionary role in the development of nearly all science and technology. Perhaps the most dramatic illustration is the development of very large-scale integrated circuits. The impact of this development on technology, our economy and quality of life cannot be overstated. The scientific and engineering challenges that had to be overcome to make this a reality were indeed tremendous, ranging from the synthesis of high-purity silicon to the exquisite materials patterning and processing techniques that permit the functional integration of semiconductors, metals and dielectrics.

In the nanoscience arena, similar or even greater integration challenges exist. Much of the fascinating behavior at the nanoscale stems from the high surface area, reduced dimensions, and unique atomic structure of nanomaterials. As these materials are coupled with other materials to form functional systems (e.g., formation of interfaces and contacts for control of active nanosystems, incorporation of nanostructures into matrix materials, introduction of heterostructured nanomaterials into new environments) it is imperative to understand how this integration affects the properties and behavior of the nanomaterials. Structural and chemical modifications at newly formed interfaces can dramatically modify the chemical, mechanical, electronic and optical properties on nanomaterials. The effects of synthesis and processing approaches on performance must be investigated and new directed and self-assembly approaches developed for greater functional control. Combined bottom-up and top-down assembly techniques must be invented to allow controlled, large-scale formation of micro-scale systems. Nanomaterials synthesis techniques must be developed that produce materials of sufficient quality and structural control that functioning and reliable systems can be formed. Finally, a detailed theoretical foundation of static, kinetic and dynamic phenomena associated with nanomaterials integration must be formed that will lead to a predicative capability for new functional materials and sytems. In order to achieve the promise of nanomaterials, these and other integration challenges must be overcome for this highly diverse class of materials.

Thus nanoscience integration extends from the synthesis and fabrication of individual nanoscale building blocks, which may in turn involve combining different materials into specific heterostructures, to the assembly of these building blocks, and finally to the generation of complex functional structures and systems. Such integration is the key to exploiting nanomaterials in applications and provides a basis for scientific investigations that will ultimately impact national and international needs in areas such as energy, environment and security.

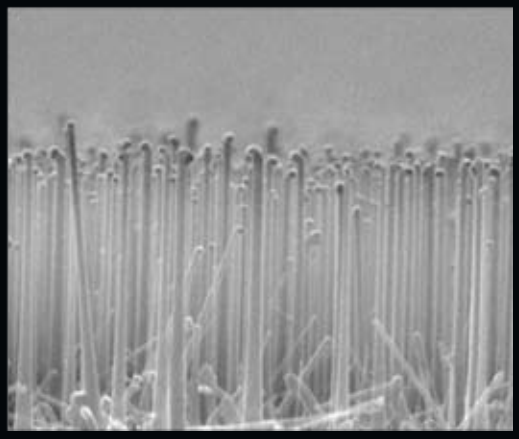
CINT was conceived to play a world-leading role to address the challenges of nanomaterials integration through synergistic coupling of our uniquely designed facilties, user program and internal science. The scientific challenges of integration demand a spectrum of synthesis, characterization and systems integration activities that span the sub-nanometer to micron length scales. The working definition for integration science is:

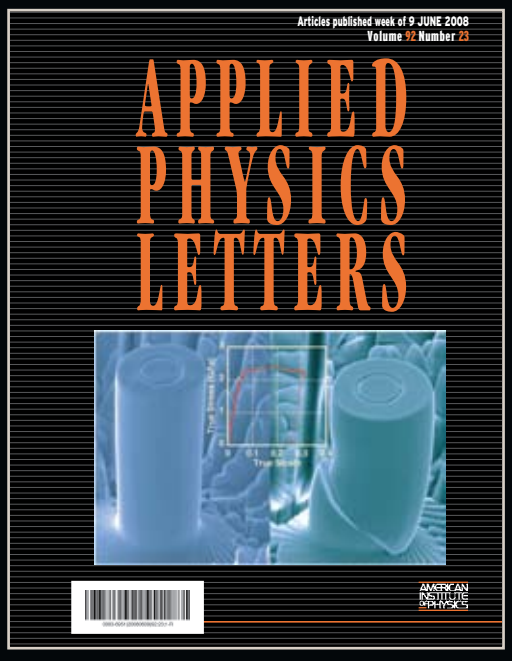
Assembling diverse nanoscale materials across length scales to design and achieve new properties and functionality.

Nanoscale integration thus extends from the synthesis and fabrication of individual nanoscale building blocks (which may combine different materials into specific heterostructures), to the assembly of these building blocks (for example as nanocomposites or patterned arrays), and finally to their incorporation into complex functional structures and systems.

One example of nanomaterials integration can be found in the area of nanowires. A growing class of materials can now be synthesized in nanowire form. This configuration enables unique control of transport related phenomena and offers many functional benefits in areas such as photovoltaics, thermoelectrics, electrical energy storage and sensors just to name a few. However, to capitalize on these unique properties, the nanowires must be integrated with other materials into systems to provide the full function required of the application. This integration poses a myriad of challenges that range from the synthesis of hybrid nanowire materials to the assembly of the optimized architecture of micro- (or macro-) scale systems. Fundamental science issues emerge such as:

- The synthesis of heterostructured nanowires of dissimilar materials (longitudinal or radial composition modulation)
- The efficiency of energy harvesting (photon, charge and/or phonon) and energy transport in nanowires and across complex interaces and how this can be controlled
- Optimizing nanowire architectures and developing assembly techniques to efficiently exploit their properties





A second example of nanomaterials integration lies in the area of nanocomposites. The integration of nanostructures into bulk composites can lead to enhanced mechancal, electrical, optical and chemical behavior. In particular, novel phenomena can be achieved by creating composites of very dissimilar materials such as semiconductor or metallic quantum dots within polymer and even biological matrixes. The integration of these materials raises a myriad of fundamental science challenges including:

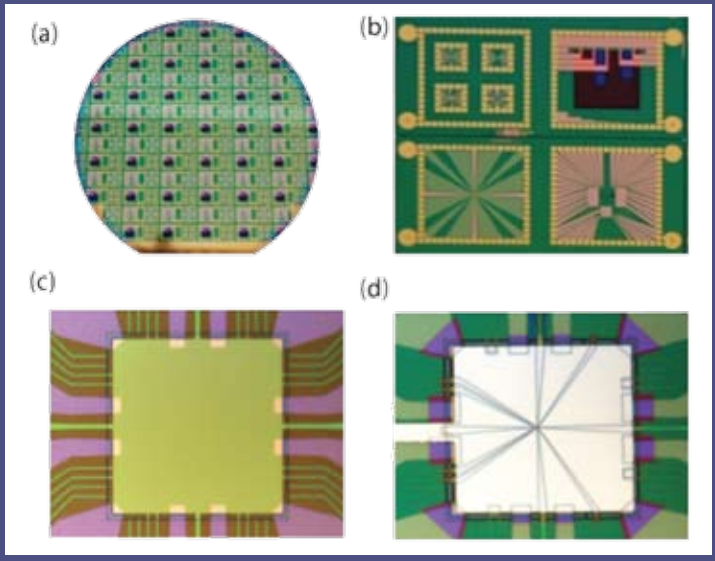
- Understanding and control of interfaces
- Energy transfer through highly dissimilar materials
- Controlled synthesis of multicomponent composites
- Characterization of embedded and complex structures and interfaces

Discovery Platforms

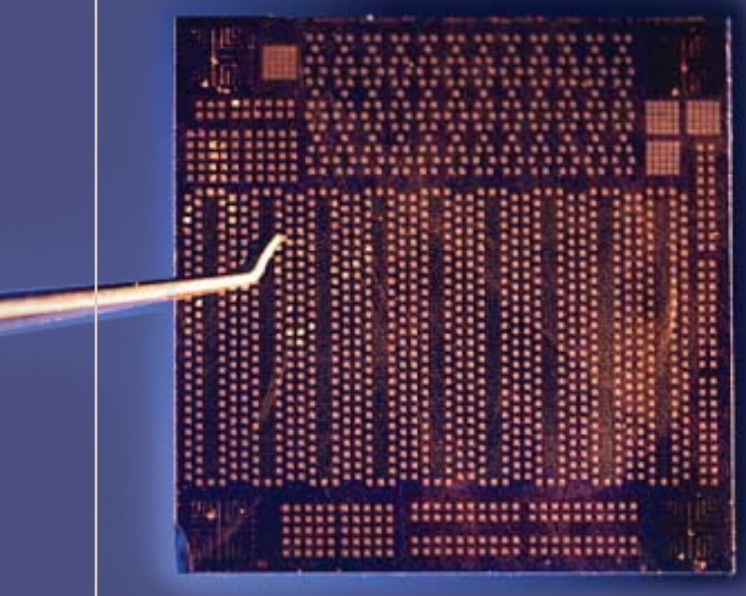
Discovery Platforms™ - The thrusts develop a variety of new instrumentation to advance the science in their areas. One effort unique to CINT is the Discovery Platform™. These platforms are modular micro-laboratories designed and batch fabricated by CINT to allow easy integration of nanomaterials into microscale structures. Their purpose is to facilitate studies of nanomaterial properties and their integration. They should allow easy connections, a range of diagnostic and experimental measurement conditions, and a degree of standardization and reproducibility in nanoscale measurements. The inception, creation and evolution of Discovery Platforms have evolved in close collaboration with our user community. The present suite of Discovery Platforms:

Electrical Transport Discovery Platform:
The Electrical Transport Discovery Platform was developed to implement metal-oxide field effect transistor (MOSFET) based nanoelectronics devices. At CINT we measure a variety of nanoelectronics devices from nanowires to patterned GaAs heterostructures to gated silicon nanostructures using this Discovery Platform.

Nanomechanics and Thermal Transport Discovery Platform:
The goal of the CINT Nanomechanics and Thermal Transport Discovery Platform is to enable researchers to perform experiments related to nanomechanics, sensing, scanning probe microscopy, in-situ TEM, and magnetization measurements, all using structures on a single, small chip-based platform. A new version of this Platform also includes structures for measurements of the electrical properties, thermal properties, electromechanical behavior, and microcalorimetry of nanoscale samples.



The Electrical Transport Discovery Platform. (a) Front-end 6 inch silicon wafer with many MOSFET structures. (b) 2 cm die with 4 quadrants for back-end nanolithography. (c) 100 micron poly gate (green) ready for depletion gate patterning. Ohmic contact to the 2D electrons are purple. (d) Same region after electron beam lithography, Al2O3 and top Al gate.



The new Nanowire Discovery Platform.

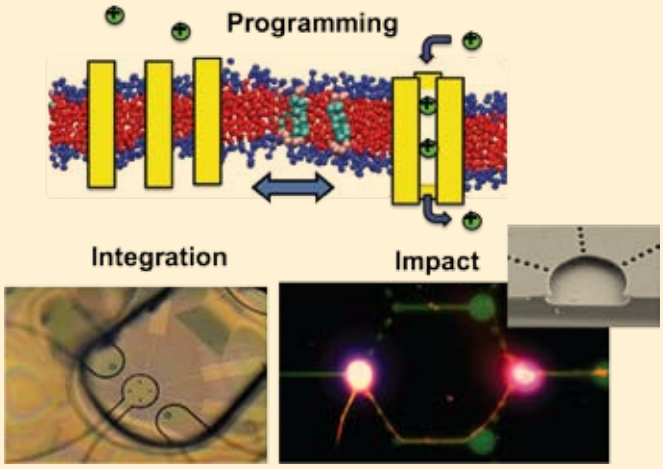
Nanowire Discovery Platform:
The Nanowire Discovery Platform is nearing completion as an experimental toolkit for nanowire and novel materials research. The NWDP builds on lessons learned in the development of the first two Discovery Platforms and will offer scientists a test bed for a diverse set of nanowire synthesis, architecture and characterization capabilities.

Integration Focus Activities

Integration Focus Activities (IFAs) are initiatives aimed at nanoscale integration challenges within the context of specific classes of materials, systems, or phenomena that will have high scientific and technological impact. These topics are developed from a "bottoms-up" process within the CINT community. They are cross-thrust activities, which provide canonical examples of integration science and are intended to become strong attractors for users and collaborators. The IFAs both emerge out of a particular thrust effort and build on areas of strength across thrusts. The studies center on particular classes of materials, systems, or phenomena and derive their integration focus through a use-inspired science approach. These focused efforts bring an interdisciplinary approach by CINT scientists and users to a particularly timely area of integration science. They enable rapid progress in specific areas of common interest, attract and build user communities, and are variable term in nature. Each IFA is supported by two dedicated postdoc positions whose research is targeted towards linking the work of CINT scientists and the user community in specific high-impact areas.

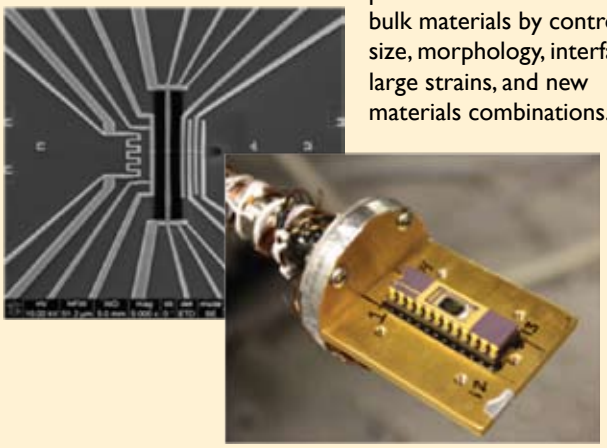
At present, two IFAs have been established:

Membrane-based Nanocomposites;
Cellular membranes enable much of the exquisite functionality that we associate with living systems. We are exploring fundamental science issues associated with developing artificial analogues to these membrane-based materials and integrating them into responsive composites and systems. We are ultimately interested in creating reconfigurable or adaptable architectures that can harvest, convert, store, and/or utilize energy sources to provide tunable and/or self-regulating photonic, electronic, mechanical, or chemical responses.



Membrane-based composites provide access to materials whose behavior mimics the multi-functional materials observed in living systems.

Nanowires for New Energy Concepts;
The novel properties in nanowires offer tremendous opportunities for transformative energy applications. CINT scientists together with our user community are focusing to understand and control the functionality and integration of heterogeneous semiconducting nanowires for new energy harvesting and storage concepts. By coupling synthesis, structure, electrical, optical, and thermal studies on specific materials systems we will bring focus to critical science questions underlying new nanowire materials concepts for photovoltaics and thermoelectrics. Heterogeneous nanowires can provide new transport properties and multi-functional performance not available in bulk materials by control of size, morphology, interfaces, large strains, and new materials combinations.



Through the Nanowire IFA, CINT scientists are trying to discover new ways to independently control thermal and electrical energy flow

User Program

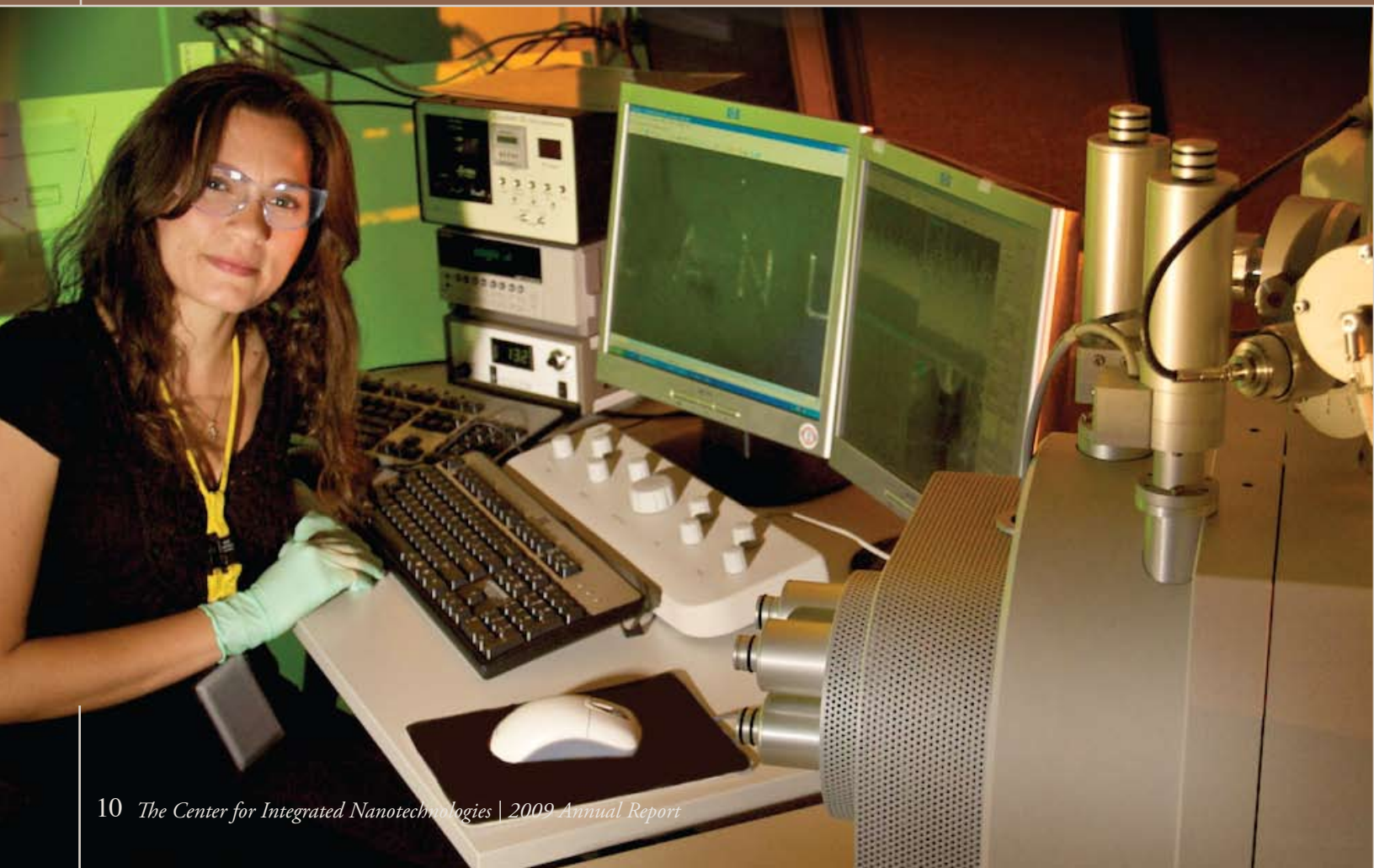
The CINT user program is designed to provide the international scientific community access to Core and Gateway Facilities. User access can include use of capabilities in either or both of our Facilities as well as engagement of the CINT Scientist staff expertise. There are two modes of user access: General User access and Partner User access, each with variable scope and the ability to conduct nonproprietary or proprietary research.

CINT users may conduct their approved research projects in collaboration with one or more CINT Scientists or may choose to access CINT capabilities independently. User proposals are evaluated on the merits of the science without preference for collaborative/independent access.

Users working independently will be properly trained and supervised by technical personnel. Some CINT capabilities cannot be operated independently for safety or complexity reasons.

User proposals consist of a 2-page (maximum) pdf document that is uploaded via the website along with user information entered on-line. All user proposals undergo an initial feasibility/safety screening by CINT technical staff, and a technical peer-review conducted by external Proposal Review Panels that reflect the four CINT Scientific Thrusts. The CINT Management uses the Review Panel scores and comments to prioritize access to CINT.

Each year, CINT and the CINT User Executive Committee (UEC) jointly organize a user meeting to highlight recent CINT user/staff research results, explore new scientific opportunities, and inform attendees about CINT capabilities and future plans. The 2009 User Meeting was held on September 29-30 in Santa Fe, NM, immediately preceding the Los Alamos Neutron Scattering Center (LANSCE) user meeting. This location enabled many of the 175 attendees to learn about both BES user facilities and tour the CINT Gateway Facility on the open afternoon between the CINT and LANSCE User Meetings. Following the successful format of the previous year, the conference began with plenary talks by Altaf Carim, DOE Office of Basic Energy Sciences, Sunil Sinha, University of California San Diego (Soft/Hard Material Interfaces), and Harry Atwater, California Institute of Technology (Plasmonics: Photons at the Nanoscale Yield Physics, Metamaterials, and Devices). These were followed by parallel mini-symposia on Nanoplasmonics: Fundamentals, Applications and Integration and Interfacial Phenomena in Nanoscience Integration. A lunchtime extended poster session enabled CINT users and scientists to present their forefront science results.



CINT Science and User Community Impact

Users by Institution

User Affiliation	2007	2008	2009
U.S. Academic	136	151	174
LANL + SNL (not CINT)	22	81	108
Other DOE Labs	3	2	10
Other FFRDC	5	6	7
Industry	6	8	20
Other	0	0	1
Foreign Institution	17	24	34
Total Users	**189**	**272**	**354**

In 2009, CINT had 412 active proposals, with users representing 37 States and 33 countries.

Our users report that their CINT research has been a significant factor in their career advancement, such as:

Margo Greenfield, hired as technical staff at Los Alamos National Laboratory
Khalid Hattar, hired as technical staff at Sandia National Laboratories
Tsing-Hua Her, promoted to tenured Associate Professor, University of North Carolina, Charlotte
Hanchen Huang, promoted from full Professor to Department Chair, University of Connecticut
Olga Kazakova, promoted to a Principal Research Scientist, National Physical Laboratory, UK
Ki-Yong Kim, hired as an Assistant Professor of Physics at University of Maryland
Junichiro Kono, promoted from Associate Professor to Professor, Rice University
Hongmei Luo, hired as an Assistant Professor at New Mexico State University
Evan Reed, hired as an Assistant Professor at Stanford University
Nian Sun, promoted to tenured Associate Professor, Northeastern University
Michael Young, hired as technical staff at Lawrence Livermore National Laboratory

Industrial User - Ed Flynn

Magnetic sensors provide early disease detection using nanotechnology

Dr. Edward Flynn, founder and president of Senior Scientific, LLC, has developed a nanotechnology-based, magnetic imaging method for early diagnosis and treatment of cancer that could provide a significant increase in detection sensitivity; much earlier detection of certain cancers; more targeted therapies; and potentially better treatment outcomes. Senior Scientific operates out of a 3,500-square-foot laboratory at the University of New Mexico's Science and Technology Park and received about $3.5 million in research grants from the National Institutes of Health since 2002 to develop this technology.

Flynn's methodology uses a superconducting quantum interference device (SQUID) sensor array to measure the magnetic fields of labeled magnetic nanoparticles that are injected into the body. The nanoparticles are attached to cells carrying specific antibodies that bind with cancer cells, and pinpoints the exact location of diseases. The sensor system can detect 50,000 cells at a distance of 4 cm. A typical mammogram needs about 10 million cancer cells for detection.

Characterization of the properties of these nanoparticles is important in order to provide proper selection of particles and optimize cell sensitivity. Facilities at the Center for Integrated Nanotechnologies are currently being used to investigate these properties. "CINT has become Senior Scientific's primary resource for characterization because of their ability to develop, characterize, and produce nanoparticles reliably," said Flynn. "Characterizing the properties of the nanoparticles involves determining the particles magnetic strength, size, and coating, using instruments that are not available at the university, and working with CINT gives Senior Scientific control of this process." Dale Huber, a principal member of the technology staff at CINT, said, "[Flynn] is a brilliant scientist and is driven by his commitment to tackling cancer. That CINT opened its labs to Senior Scientific reflects the caliber of Flynn's achievements."

In 1998, after Flynn's second wife was diagnosed with breast cancer, Flynn began his research using magnetic particles to detect cancer cells. Flynn said, "If we can detect cancer much earlier before it spreads, there is higher likelihood for successful

From Discovery to Innovation…

treatment." Flynn's technology could lead to more targeted treatment because the method detects precisely where a cancer is located. Senior Scientific's diagnostic methods include the detection and localization of breast cancer, ovarian cancer, leukemia, rejection of transplanted organs, and Alzheimer's Disease.

Flynn's medical diagnostic breakthrough has grabbed the attention of high-tech investment firm Manhattan Scientifics Inc., leading to an agreement with Flynn and his company to acquire all the manufacturing and marketing rights, together with all commercial rights associated with Flynn's patents and intellectual property in the emerging field of nanomedicine. Marvin Maslow, chairman emeritus of Manhattan Scientifics, said, "It is not merely our goal, but our obligation to bring Dr. Flynn's extraordinary work in the field of early cancer detection and potential treatment to the world medical community. Just as we have demonstrated in the past, we intend to identify one or more appropriate Fortune 500 industrial partners in the pharmaceutical and medical device industries to bring product to market."

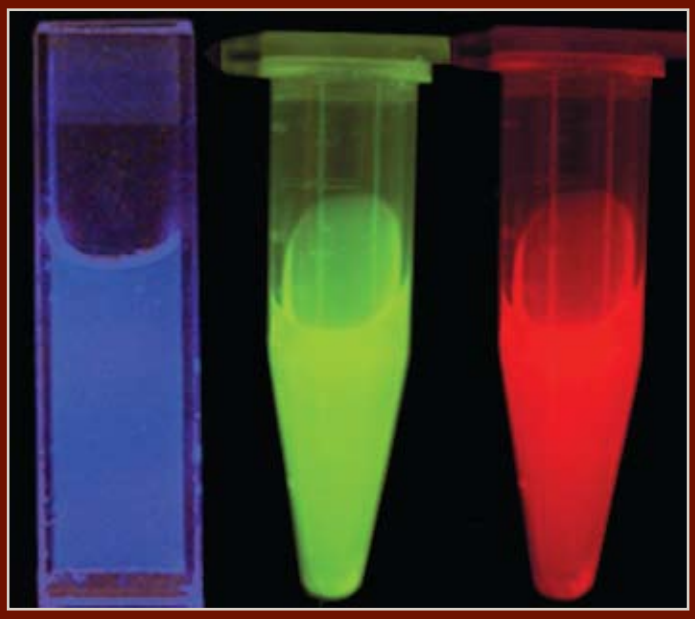
"Translating research into development by demonstrating that a method can be commercially marketed and used clinically is a major goal and challenge of the program," said Flynn. "I am confident that partnering with Manhattan Scientifics will achieve this goal, and bring the product to the medical community."

Flynn, a Los Alamos National Laboratory (LANL) Fellow, worked at LANL for 38 years, publishing 185 scientific papers in the field of nuclear physics. After the death of his first wife, Flynn began his research into medical physics, where he launched the Biophysics group, initiated the Brain Research Program at LANL, published 65 scientific papers, and was the founding Director of the Mind Research Network. Thereafter, he founded Senior Scientific, a privately-held company, pioneering his important cancer research.

For Flynn the primary reward for his cancer research would be, "To see it working; that research moves to development and becomes reality in the clinical environment. My ultimate reward is in helping people."

Scientific discovery inspires the innovation that drives economic prosperity – but discovering new properties of nanometer-size materials is insufficient to ensure technological innovation and benefit to humankind. This is why CINT focuses on integration issues. What knowledge is needed in order to exploit nanomaterials for various applications?

By attracting researchers interested in these problems, CINT creates scientific communities that tackle these challenges. In fact, virtually all of CINT's user projects involve teams of researchers: consisting of researchers from academia, research laboratories or private-sector companies working with CINT Scientists. Approximately half of the user projects involve multiple CINT Scientists, thereby pulling together the combined expertise that cannot be found in one institution.



CINT accepts hundreds of user-defined projects each year, generating knowledge that ranges from the most basic physics to valuable intellectual property protected by patents. Although the majority of the user projects involve pre-competitive research that will be published in the peer-reviewed technical literature, CINT users can conduct proprietary research as well. User proposals containing proprietary information are reviewed via a separate process to maintain confidentiality under protection of an executed Non-Disclosure Agreement between CINT (Los Alamos National Laboratory and Sandia National Laboratories) and the prospective user's institution.

CINT also recognizes that the pace of innovation often requires rapid investigation. Hence, CINT accepts "Rapid Access" user proposals, submitted via the web site, that clearly demonstrate a need for immediate access to conduct well-focused, short-term work with extremely high-impact potential. If approved, Rapid Access User Projects remain valid only until the beginning of the next available regular application cycle.

For further information about private-sector research at CINT (either pre-competitive or proprietary), please contact the CINT User Program Manager, Neal Shinn (ndshinn@sandia.gov).

Facilities

CINT facilities are the CINT Core Facility, located in Albuquerque, NM, and operated by Sandia National Laboratories and the Gateway to Los Alamos Facility, located in Los Alamos NM and operated by Los Alamos National Laboratory. The co-location of these two dedicated facilities enables CINT users to benefit from other capabilities at both Laboratories, including the Lujan Neutron Scattering Center, National High Magnetic Field Laboratory, and leveraged capabilities at LANL.

Through these two facilities, CINT has thirteen major capabilities available to scientists and users.

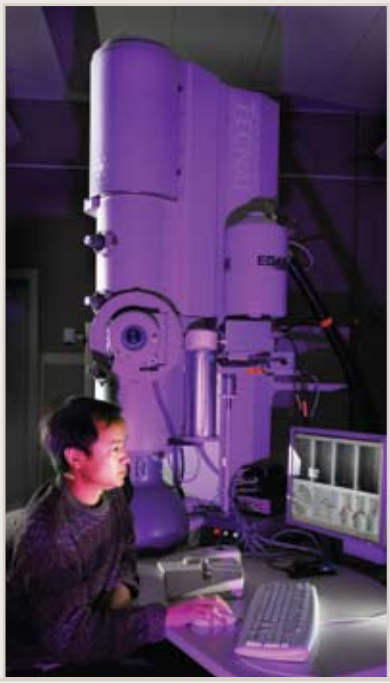
1. Transmission Electron Microscopy
2. Molecular Beam Epitaxy
3. Integration Laboratory
4. Electrom Beam Lithography
5. Quantum Transport
6. Laser-Based Spectroscopies
7. Scanning Probes & Imaging
8. Nanomechanics
9. Discovery Platforms
10. Materials Synthesis
11. Bio-Materials
12. Optical Microscopy
13. Computation and Visualization

Science Thrust Descriptions

Integration is the key to exploiting the novel properties of nanoscale materials and subsequently creating new nanotechnologies to benefit society. Hence, the CINT scientific community is built around nanomaterials integration. The scientific staff and capabilities at CINT are organized into four interdisciplinary Science Thrusts:

Nanoscale Electronics and Mechanics

The focus is on understanding and controlling electrical and mechanical properties arising from confinement on the nanoscale and interactions within nanostructures. Significant challenges in integration science issues include:



1) nanowire integration science for new energy concepts;
2) manipulation of spin properties of single electrons in semiconductors;
3) surface and interface driven mechanics of nanoscale materials;
4) understanding structure-property relationships in nanoscale materials; and
5) developing multi-functional composite materials exploiting electronic, mechanical and magnetic interactions.

Nanophotonics and Optical Nanomaterials

The emphasis is on understanding and controlling fundamental photonic, electronic, and magnetic interactions in nanostructured optical materials. Key integration science challenges include:

1) chemical and physical synthesis of optical, electronic, and magnetic nanomaterials;
2) collective and emergent electromagnetic phenomena (plasmonics, metamaterials, photonic lattices, solitons);
3) multifunctional behavior in hybrid nanostructures comprising optical components; and
4) energy transformations on the nanoscale.

Soft, Biological, and Composite Nanomaterials

This thrust concentrates on solution-based, "bottomup" approaches for development of integrated nanomaterials. The emphasis is on key scientific areas that underlie the challenge of nanoscience integration. These include:

1) controlling interfaces and their interactions;
2) developing new characterization tools that provide information on multiple length and time scales; and
3) exploring the roles of disorder and dynamics in determining materials performance.

Theory and Simulation of Nanoscale Phenomena

Scientists in this thrust develop analytical and computational approaches that enable the calculation of the competing interactions and the resultant material structure and properties that occur in integrated nanomaterials systems.

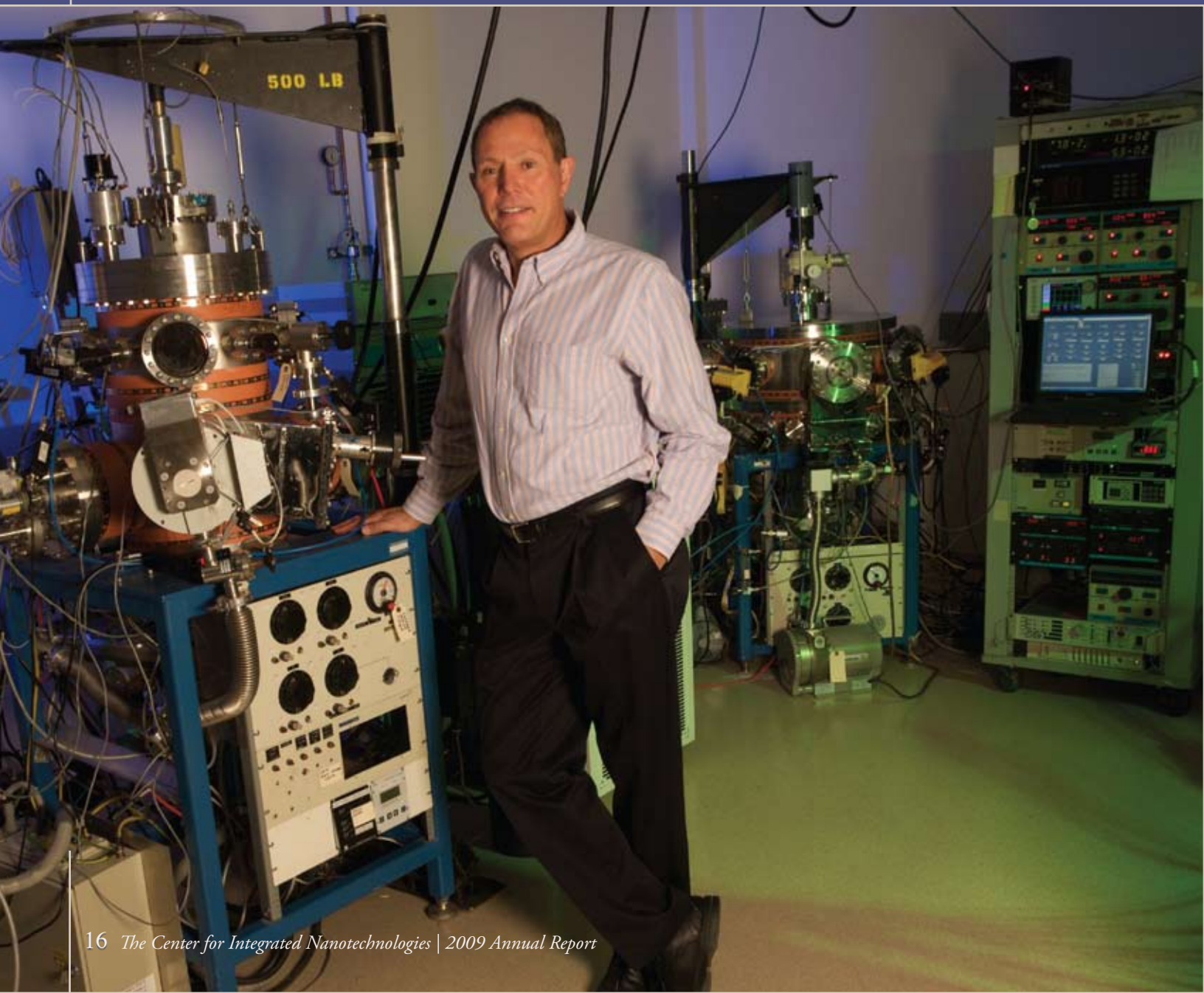
The strategy emphasizes the development and application of advanced techniques drawn from combinations of many-body, local density, molecular dynamics and other methods capable of describing complex materials.

Nanoscale Electronics and Mechanics

The Nanoscale Electronics and Mechanics (NEM) thrust focuses on the electronic and mechanical properties of nanosystems and issues related to integration of a wide variety of nanoscale materials. Ongoing research includes plastic deformation, elastic and fracture properties on the nanoscale, coherent transport and interactions of low dimensional systems, coupled mechanical systems, coupling of mechanical and electronic properties, structural and electronic properties of nanowires, and investigation of materials interface properties. This research will be strongly supported by an effort in nanofabrication with an emphasis on integration of electrical and mechanical systems in addition to integration with composite nanomaterials and biological systems. Unique tools in this thrust include molecular beam epitaxial growth of semiconductor heterostructures for producing ultra-clean low dimensional electron systems, a high current state of the art ion implanter, new tools for nano-manipulation, in situ STM/TEM and specialized growth techniques for films, nanowires and other nanostructures. This thrust is developing two Discovery Platforms™. The Cantilever Array Platform will provide mechanical tools optimized for nanoscience experiments. The Electrical Transport and Optical Spectroscopy Platform will enable reliable, high throughput electrical and optical measurements compatible with CINT fabrication and characterization equipment.

Scientific directions include:

- Control of electronic transport and wavefunctions using nanostructured materials,
- Mechanical properties and coupling of nanostructured materials,
- Exploration of new ways to integrate diverse classes of functional materials on the nanoscale.

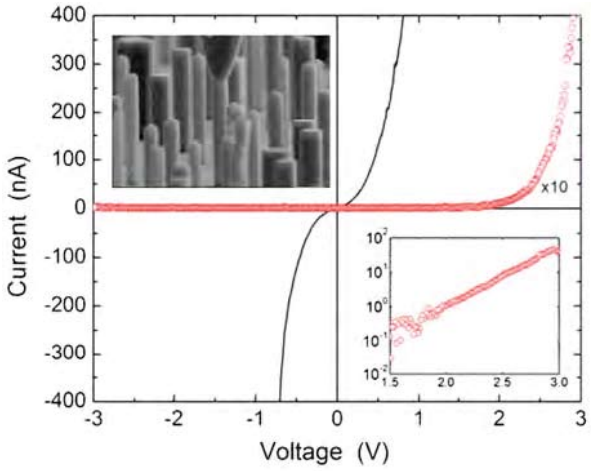


Unusually Strong Space-Charge-Limited Current in Thin Wires

Accomplishment: The current-voltage characteristics of thin semiconductor wires are often observed to be nonlinear – often attributed to Schottky barriers at the contact interface. Our electronic transport measurements of GaN nanorods demonstrate that the nonlinear behavior originates instead from space-charge-limited current due to the high aspect ratio of these nanostructures.

We use a CINT-built nanomanipulator inside of a Scanning Electron Microscope (SEM) that can position a very sharp probe with nanometer precision. The SEM is used to see the nanorods and guide the probe into contact with selected rods. Once contacted, an electric current passing through the probe and the rod measures the electronic characteristics of the rod and the interface. We find that a Schottky barrier can indeed be formed at the probe/rod interface, but this interface barrier is easily broken down to measure the space-charge-limited character of the rod itself. Space-charge-limited current is unusually strong in nanorods because of their high aspect ratio and their inherent large surface-to-volume ratio. We developed a theory of space-charge-limited current in thin wires that corroborates the experiments and shows that poor screening in high-aspect ratio materials leads to a dramatic enhancement of space-charge limited current, resulting in new scaling in terms of the aspect ratio.

Significance: Nanowires made from a variety of semiconducting materials are showing great promise as active elements in electronics and are being extensively characterized electrically. However, transport properties of nanoscale contacts can differ significantly from their bulk counterparts. Our goal is to understand transport in thin wires, the origin of diode characteristics at nanoscale metal-semiconductor contacts, and the performance of engineered junction diodes in nanowires so that individual nanowires can be predictably and reliably integrated into unique device structures.



Current-voltage characteristics before (symbols) and after (line) the tip is pressed into the nanowire. The bottom right inset shows the current before pressing on a log scale. The top left inset is a SEM image of the Au-coated W tip contacting a GaN nanowire.

Attribution: User Project Number U2008A071 Characterization and Device Integration of Vertical GaN Nanorods
Publications:

"Unusually Strong Space-Charge-Limited Current in Thin Wires", A. A. Talin, F. Léonard, B. S. Swartzentruber, X. Wang, and S. D. Hersee, Phys. Rev. Lett., 101(7), 076802 (2008).

"GaN Nanowire Light Emitting Diodes Based on Templated and Scalable Nanowire Growth Process", S. D. Hersee, M. Fairchild, A. K. Rishinaramangalam, M. S. Ferdous, L. Zhang, P. M. Varangis, B. S. Swartzentruber, and A. A. Talin, Electronics Letters, 45(1), 75 (2009).

"Electrical Transport in GaN Nanowires Grown by Selective Epitaxy", A. A. Talin, B. S. Swartzentruber, F. Léonard, X. Wang, and S. D. Hersee, J. Vac. Sci. Tech. B, 27(4), 2040 (2009).

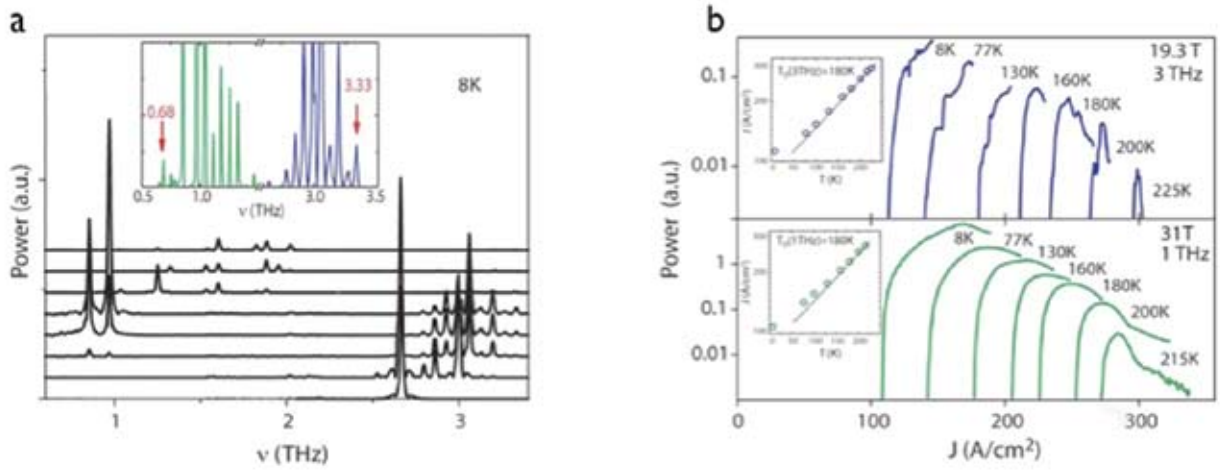
"Electronic Transport in Nanowires: From Injection-Limited to Space-Charge-Limited Behavior", F. Léonard, A. A. Talin, A. M. Katzenmeyer, B. S. Swartzentruber, S. T. Picraux, E. Toimil-Molares, J. G. Cederberg, X. Wang, S. D. Hersee, and A. Rishinaramangalum, SPIE, 7406(1), 74060G (2009).

Magnetic field assisted multi-wavelength (sub)THz quantum cascade laser operating up to 225K

Accomplishment: Advances in semiconductor bandgap engineering have resulted in the recent development of the THz quantum cascade laser (QCL). In QCLs, radiative transitions take place between size-quantized subbands within the conduction band of a multi-quantum well (QW) system. In this way, the QW and barrier thicknesses can be varied allowing the emission wavelength to be tailored using the same material in order to cover a broad range of frequencies. The epitaxial layer thicknesses typically range from 3 to 35 monolayers (1.5 to 10nm) and are stacked to a total thickness of about 10μm.

These compact, optoelectronic devices are now covering the frequency range from 1.2 THz to 5 THz, though cryogenic cooling is still required. Further progress towards the realization of devices operating at higher temperatures (eventually at room temperature) and emitting at longer wavelengths (sub-THz QCLs) becomes ever more difficult because it requires maintaining a population inversion between closely-spaced electronic subbands (1THz ~ 4meV). We have demonstrated that a magnetic field assisted QCL, based on the resonant-phonon design, can achieve laser emission from a single device in a wide range of frequencies (0.68THz to 3.33THz) by applying the appropriate electrical bias and magnetic field. In addition, the device shows 1THz laser action at temperatures up to 215K, and 3THz lasing up to 225K.

Significance: Quantum cascade lasers demonstrate integration over a wide range of length scales. Individual atomic layers at a thickness of less than a nanometer are carefully stacked to control the electronic structure of the new material. The total thickness of the stack is measured in microns, which is then processed into a laser with dimensions on the order of a few millimeters. The package laser can then be inserted into operating sensing systems.



QCL device performance in terms of spectral coverage and operational temperature. (a) Spectral coverage of the QCL device with increasing voltage bias and magnetic field (bottom curve 54.9mV/period, 13T; top curve 88.4mV/period, 25T). The inset shows the QCLs spectral extremes: 0.68THz (69.9mV /period, 31.2T) and 3.33THz (63.9mV/period, 19T). (b) Temperature dependence of P(J) at two enhanced lasing positions at 19.3T and 31T. The inserts show the current density threshold as a function of the temperature. 1THz lasing has been omitted from the 19.3T curves for visual clarity.

Attribution: User Project Number U2008B020

Publications:

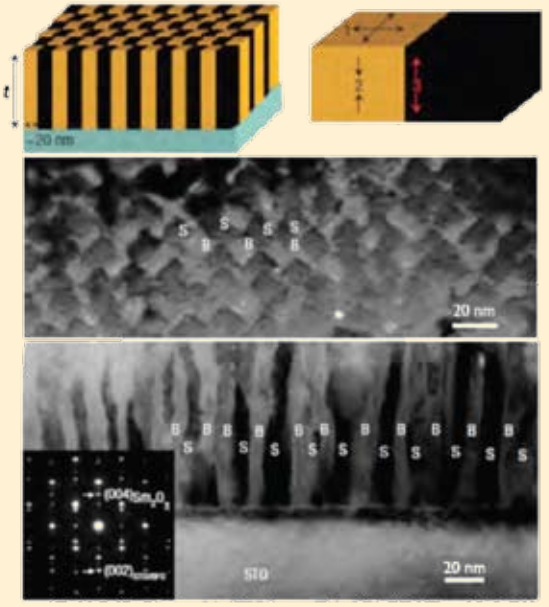
Wade, A., Fedorov, G., Smirnov, D., Kumar, S., Williams, B.S., Hu, Q., and Reno, J.L., Magnetic field assisted multi-wavelength THz quantum cascade laser operating up to 225K, Nature Photonics, 3, January 2009.

Strain induced phenomena in vertically aligned nanocomposite films

Accomplishment: Over the past several years, new discoveries and major advances have been made to enhance the understanding of metal-oxides and their properties such as magnetism, ferroelectricity, and superconductivity. More recently, new multifunctionalities have been achieved through interfacing different oxides at the nanoscale by forming nanocomposite films. The emergent behaviors in composite films that cannot be obtained in the individual bulk constituents provide a new material design paradigm to control and manipulate the physical properties of such composite films and produce novel functionalities.

We have used an innovative approach to control the strain states in nanocomposite films (both BiFeO3:Sm2O3 and LSMO:ZnO) by forming spontaneously ordered pillar structures (see Figure). We have made significant discoveries: 1) the columns control the strain in one another in the vertical direction, independent of the lateral interface; 2) the strain is maintained in rather thick films (the thickest film we have studied is 450 nm and the strain still persists); These advances allow an order of magnitude thicker strained films to be grown relative to lateral systems, enabling vast improvement of properties such as the dielectric loss for BiFeO3.

Significance: The degree of strain control holds great promise for novel lattice-engineered functional property control that goes substantially beyond what is possible in lateral strain control as used for in established systems. The work also opens up a new avenue for strain control in relatively thick films so that materials with, for example, dielectric or optical applications can be tuned by the appropriate choice of second phase. The nanostructures spontaneously ordered over large areas have important ramifications for many areas of functional device materials.



Strain concept and TEM images of the BFO:SmO nanocomposite film in a heteroepitaxial vertical nanocomposite. (Top) schematic illustration of the strains present in vertical nanocomposites: arrows show the strains along different directions; (Middle) :pPlan-view showing an ordered checkerboard structure (B represents BFO, S represents SmO); (Bottom) low magnification cross-sectional view showing the alternating columns of BFO and SmO.

Publications:

J. L. MacManus-Driscoll, P. Zerrer, H. Wang, H. Yang, J. Yoon, S. R. Foltyn, M. G. Blamire, and Q. X. Jia, "Spontaneous Ordering, Strain Control and Manipulation in Vertical Nanocomposite Heteroepitaxial Films," Nature Materials 7, 314 (2008).

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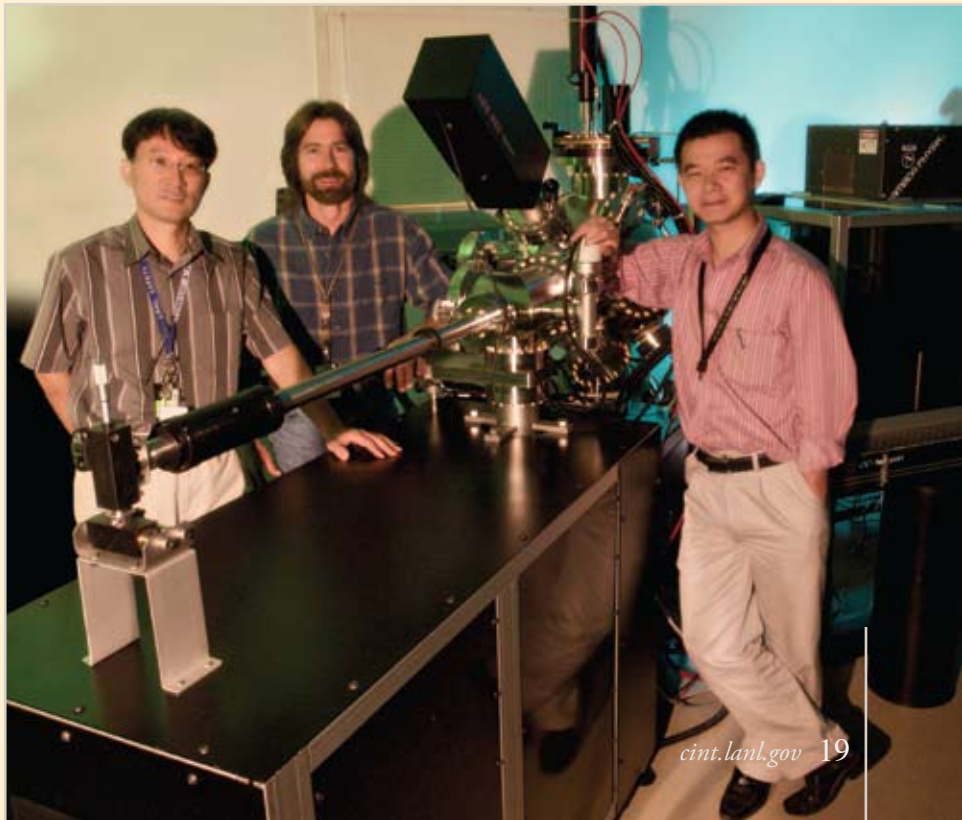
Z. Bi, J. W. Lee, H. Wang, H. Yang, Q. X. Jia, and J. L. MacManus-Driscoll, "Tunable Lattice Strain in Vertically Aligned Nanocomposite (BiFeO3) x(Sm2O3) 1-x Thin Films," J. Appl. Phys. 106, 094309 (2009).

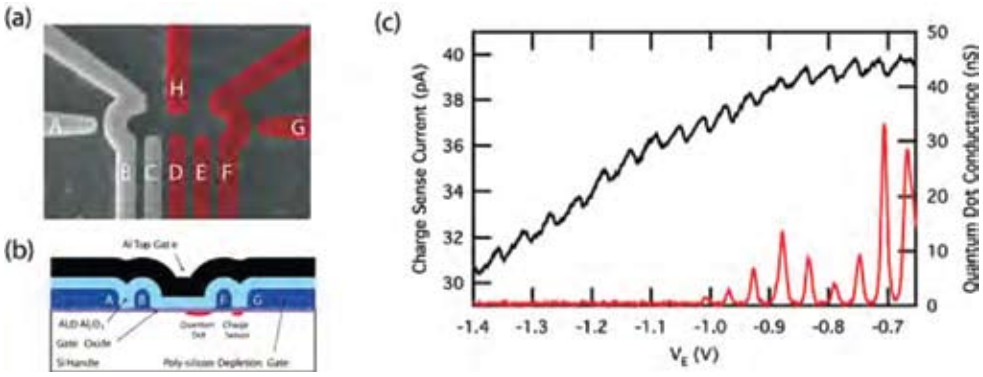
A. Fouchet, H. Wang, H. Yang, J. Yoon, Q. X. Jia, and J. L. MacManus-Driscoll, "Spontaneous Ordering, Strain Control and Multifunctionality in Vertical Nanocomposite Heteroepitaxial Films," IEEE Trans. Ultrasonics, Ferroelectrics, and Frequency Control 56, 1534 (2009).

H. Yang, H. Wang, J. Yoon, Y. Q. Wang, M. Jain, D. M. Feldmann, P. C. Dowden, J. L. MacManus-Driscoll, and Q. X. Jia, "Vertical Interface Effect on the Physical Properties of Self-assembled Nanocomposite Epitaxial Films," Adv. Mater. 21, 3794 (2009).



Charge sensing in silicon metal-oxide-semiconductor quantum dots

Accomplishment: Silicon MOS nanostructure devices are fabricated using a version of the CINT Electrical Transport Discovery Platform. In the Microelectronics Development Lab, silicon wafers are fabricated with a stack that included ohmic contacts, a high quality thermal oxide, a doped poly-silicon layer and a region for additional nanolithography (Figure 1b). In order to operate sub-100 nm structures, the poly-silicon gate is patterned using electron beam lithography and subsequent dry etching. A negative voltage expels electrons from the depletion gate regime and defines the nanostructure. Field-effect based devices also require a positive gate voltage to induce electrons at the silicon-silicon oxide interface, and this is implemented using a separate overall Al top gate. The top gate and depletion gates are electrically isolated with atomic layer deposition of aluminum oxide.



a) Double quantum dot is defined by depletion gates (B-F and H); integrated electrometers are the point contacts defined by A-B and F-G. (b) Cross section of double gated MOSFET layers with the dot and point contacts represented schematically. (c) Coulomb blockade peaks in a many-electron single dot (defined by H,D,E and F). The compensated charge sensor current shows a step each time another electron is added to the dot.

The device shown in the Figure is designed as a double quantum dot (gates B-F,H) with narrow constrictions to the left and right of the double dot regime (gates A-B and F-G). In future experiments we plan to use the full double dot structure, but for the charge sensing demonstration we use the gates highlighted in red to form a single quantum dot (D-F and H) with an integrated charge sensor (F-G). The quantum dot has approximately 100 electrons, and a negative bias on the plunger gate, E, reduces the electron number of the dot by a single electron every 43 mV. The electron addition can be observed directly by monitoring the transport through the dot (Figure 1c, red line). Each time an electron energy level aligns with the reservoirs, the current increases and when the levels are not aligned the dot conductance is blocked. The point contact constriction can be used as a remote sensor for the changes in electron number (Figure 2c, black line). For a fixed bias, the current changes by approximately 3% every time the electron number in the dot changes. The charge sensor can effectively measure changes in electron occupation even when the direct conductance through the dot is too small to observed Coulomb blockade (VE<-1 volt)

Significance: Spin quantum computing in semiconductors requires working with single electrons in quantum dots. In this regime it is important that no current flow through the dot. The successful demonstration of a point contact electrometer in a silicon MOS nanostructure is a critical element to both reducing the electron number to one, and eventually for measuring the spin using a spin to charge transduction technique. This approach is also compatible with sub-microsecond single electron measurements using an rf-SET approach.

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Nordberg, E.P., et al., Charge sensing in enhancement mode double-top-gated metal-oxide-semiconductor quantum dots. Applied Physics Letters, 2009. 95(20): p. 202102.

Nordberg, E.P., et al., Enhancement-mode double-top-gated metal-oxide-semiconductor nanostructures with tunable lateral geometry. Physical Review B, 2009. 80(11).

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Nanophotonics and Optical Nanomaterials

The Nanophotonics and Optical Nanomaterials (NPON) thrust addresses the overall scientific challenge of understanding and controlling fundamental photonic, electronic and magnetic interactions in nanostructured optical materials fabricated using both chemical and physical syntheses.

Ongoing research includes colloidal synthesis of semiconductor, noble-metal and magnetic-metal nanostructures having controlled shape (anisotropy) and surface chemistry (reactivity), as well as hybrid, multifunctional (e.g., magneto-optical, electro-optical, and multi-ferroic) nanomaterials comprising semiconductors and metals. Bottom-up assembly approaches (e.g., self assembly, colloidal lithography, layer-by-layer deposition, and directed/field- or interface-mediated assembly) are used to prepare active sol-gel and polymer nanocomposites and synthetic opals. Further, polymer-assisted thin-film growth techniques and pulsed laser deposition are used to grow synthetically challenging complex metal oxides. Lithographic methods are applied to the fabrication of two- and three-dimensional photonic crystals, while physical synthesis (e.g., MBE, VLS) is used to fabricate epitaxial quantum dots, quantum well structures, and quantum wires.

In an effort to explore new properties stemming from the creation of novel interfaces and hybrid structures, we focus on developing new materials that bring together disparate material types for creating coupled or additive behavior in addition to multifunctional properties. Here, we emphasize the interface between semiconductors and metals, inorganics and organics, and colloidal and epitaxial. Finally, for both the synthesis and assembly of optical nanomaterials, we utilize the CINT Electrical Transport & Optical Spectroscopy Discovery Platform™ and the Microfluidics Synthesis Platform. The preparation and fabrication of such novel nanomaterials allows us to study and control collective and emergent electromagnetic phenomena.

We specifically explore the areas of nanophotonics, plasmonics, metamaterials (left-handed and photonic crystals), and solitons. Further, we use our unique capabilities in advanced ultrafast and single-nanostructure spectroscopies, including various scan-probe techniques, to explore energy transformations on the nanoscale from energy and charge transfer to electronic relaxation across multiple length-, time-, and energy scales. Such phenomena will contribute to "light capture" applications (photovoltaics, photodetection, and radiation detection), as well as to "light emission" applications (solid-state lighting, LEDs, lasing, etc.).

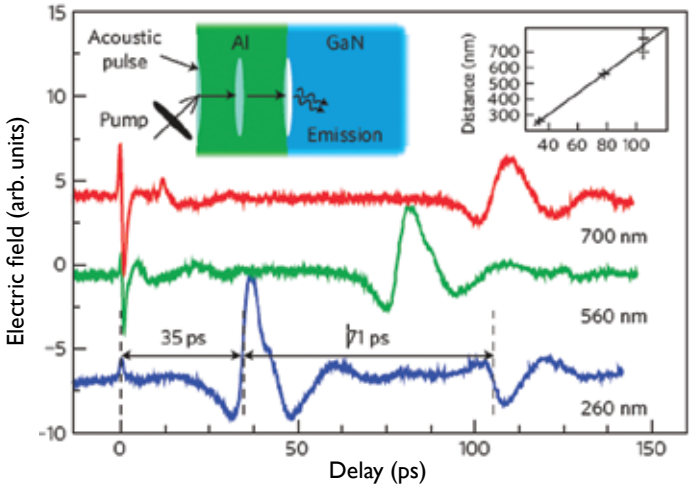
Scientific directions include:

- Chemical synthesis of optical, electronic, and magnetic nanomaterials
- Physical synthesis and fabrication of optically active nanomaterials
- Collective and emergent electromagnetic phenomena (plasmonics, metamaterials, photonic lattices, solitons)
- Multifunctional behaviors and hybrid nanostructures
- Optical excitation and energy transformations on the nanoscale

Observation of terahertz radiation coherently generated by acoustic waves

Accomplishment: Experiments using subpicosecond lasers have demonstrated the generation and detection of acoustic and shock waves in materials with terahertz frequencies leading to new techniques for probing the structure of thin films. However, most existing approaches to detect and measure the time dependence of terahertz-frequency strain waves in materials use time-resolved interferometry or reflectrometry. Here, we describe results demonstrating acoustically generated terahertz radiation that is coherently related to the strain-wave time dependence.

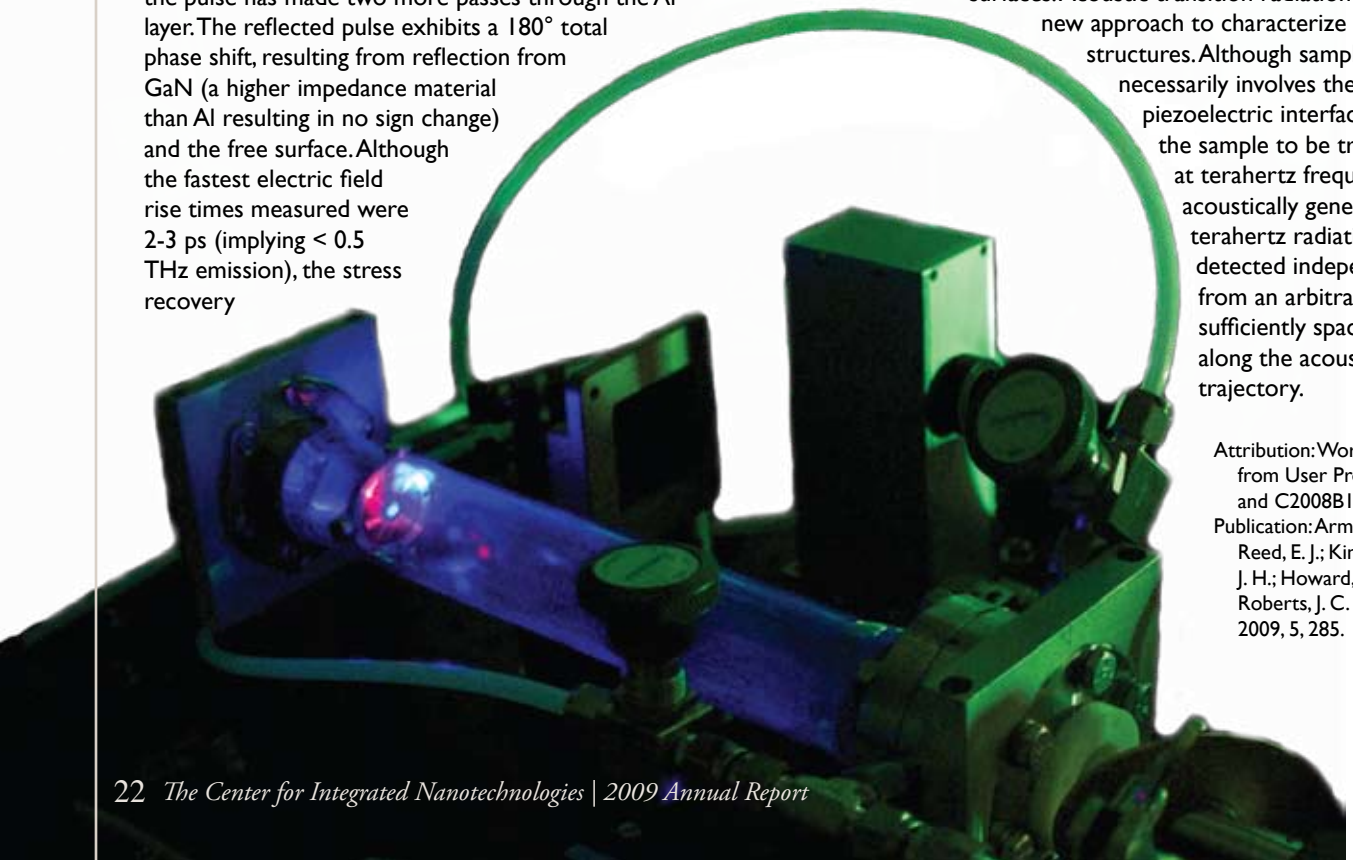
In contrast to commonly used terahertz generation, we report observations of radiation generated by the terahertz-frequency acoustic wave; the signal is not conversion of an ultrashort optical pulse to terahertz radiation through the optical response of the sample. Boundaries between regions of differing piezoelectric response are formed by a submicrometre thick layer of aluminium on gallium nitride, and a layer of AlN embedded in the Ga. The acoustic wave is generated by focusing an ultrashort (~100 fs) optical pulse onto the Al layer, which heats the metal within ~ 3.5 ps through a depth of about 50 nm. Thermal expansion generates a wave with maximum strain of the order of -0.01 corresponding to pressures of the order of 1 GPa. The figure shows the detected electric field for various thicknesses of Al coated on GaN. The electric field varies corresponding to the acoustic pulse arrival at the Al-GaN interface. The arrival times at the interface vary linearly with the thickness of the Al layer in each sample (see figure inset), demonstrating a linear correlation between the time of emission and the acoustic transit time with a fitted acoustic velocity of 6.7 km/s (compare to 6.4 km/s sound speed in Al). The figure also shows the first reflection from the Al-GaN boundary and the Al-free surface for the thinnest sample, after the pulse has made two more passes through the Al layer. The reflected pulse exhibits a 180° total phase shift, resulting from reflection from GaN (a higher impedance material than Al resulting in no sign change) and the free surface. Although the fastest electric field rise times measured were 2-3 ps (implying < 0.5 THz emission), the stress recovery



Electric field from Al–GaN interface versus time labeled by Al layer thickness. Dispersion in the shape of the pulse is primarily due to polycrystalline effects in the Al layer. An acoustic reflection in the Al layer radiates around 105 ps in the 260nm Al-coated sample. The transit times are labeled for the 260nm trace. Inset: Layer thickness versus transit time in picoseconds.

exhibits lower acoustic bandwidth. For instance, the fast, expansive portion of the stress in the 260nm Al sample has a 5 ps rise (compared with a ~3 ps rise in the electric field), consistent with acoustic measurements in related systems using interferometry.

Significance: The term `nanoseismology' has been proposed to characterize the study of thin-film nanostructures using high-frequency acoustic waves. Such studies so far have used optical techniques to observe reflected strain waves at surfaces. Acoustic transition radiation enables a new approach to characterize piezoelectric structures. Although sample preparation necessarily involves the fabrication of piezoelectric interfaces and requires the sample to be transmissive at terahertz frequencies, acoustically generated terahertz radiation may be detected independently from an arbitrary number of sufficiently spaced interfaces along the acoustic wave trajectory.



Attribution: Work derived in part from User Project R2007A068 and C2008B102.
Publication: Armstrong, M. R.; Reed, E. J.; Kim, K.-Y.; Glownia, J. H.; Howard, W. M.; Piner, E. L.; Roberts, J. C. Nature Physics 2009, 5, 285.

CINT Scientist Highlight - Aaron Gin

Like any good photographer, Aaron Gin must know how to balance light and exposure when taking high-quality pictures. Fortunately for this CINT scientist, his knowledge of proper photography principles applies not only to his hobby shooting portraits but also to his CINT projects fabricating nanoscale materials and devices.

"My passion for photography has a lot of parallels with my science," says Gin, who splits his time between CINT and the Photonic Microsystems Technologies department at Sandia National Laboratories.

Gin started work at Sandia four years ago, after his doctorate in electrical engineering at Northwestern University. Upon arrival, he began work on superlattice infrared detectors, which take advantage of alternating, nanometer-thick layers of semiconductors like indium arsenide and gallium antimonide. Within a year, Gin joined CINT to collaborate with others, primarily in the area of nanophotonics and optical nanomaterials.

Creating the proper exposure for a photograph, Gin says, is analogous to making the correct exposure in a nanofabrication process called electron beam lithography. Instead of channeling the proper amount of light at the right shutter speed through a camera, he focuses a few nanometer-diameter electron beam of highly energetic electrons onto nanomaterials to form a specific "picture" or pattern on a substrate. These exposed patterns are chemically revealed in a process similar to traditional darkroom film development. Finally, the nanometer-scale patterns can be transferred onto the substrate using a deposition, etch, or metal liftoff. Through iteration and carefully adjusted process steps, Gin says, a scientist can fabricate a nanoscale device.

Gin says he appreciates how CINT draws scientists from both Los Alamos and Sandia National Laboratories to work together to further the revolutionary field of nanoscience integration.

"You end up with a sort of scientific All-Star team, folks not only good at what they do in their own technical work, but they're also very good collaborators," he says.

At CINT, Gin leads the design and fabrication of the Nanowire Discovery Platform, a experimental testbed that allows researchers to study nanowires, or long, "thin" structures (also called 1-D nanostructures) of less than 100 nanometers in diameter. By enabling research on nanowires and graphene, as well as their novel transport properties, "we hope to realize the promise of new semiconductor materials and devices," Gin says.

His colleagues value how Gin's experience in optoelectronic devices brings fresh perspectives into CINT's collaborative projects.

"This is especially beneficial for semiconductor nanostructure devices, since complex fabrication processing steps involving three-dimensional hybrid integration of dissimilar materials are often employed," says Ping-Show Wong, a University of California at Los Angeles (UCLA) postdoctoral scholar. "Aaron's keen diagnostic ability and capabilities to resolve scientific problems have been proven exceptional in our many years of collaboration."

Gin's work at CINT with Wong and the group led by Prof. Diana Huffaker of UCLA's Electrical Engineering Department involves the material synthesis, characterization, and device realization of semiconductor nanowires grown on nanopatterned substrates. The project deals with the fabrication of both individual and arrayed nanowire electrical and optoelectronic devices that can be used for transistors and solar cells.

Gin's colleagues also appreciate his ability to communicate well and to ask probing questions to in order to refine his contributions.

"Aaron brings a great deal of skill and insight to his work. He works efficiently and provides rapid turnaround even when dealing with advanced cleanroom processing techniques," said Paul Davids, a project team member at CINT and scientist in Sandia's Applied Photonics and Microsystems group. Gin's work with Davids led to prototype devices and experiments, "which have led to a great deal of excitement internally to Sandia," Davids said.

That excitement continues to infuse the work and collaborations of Gin and his colleagues, Gin says. "If you talk to any other CINT scientist, I think they'll have a level of excitement similar to mine. We understand that we're on the cutting edge, and we have the facilities, resources, and scientific collaborations to make progress towards these great opportunities."

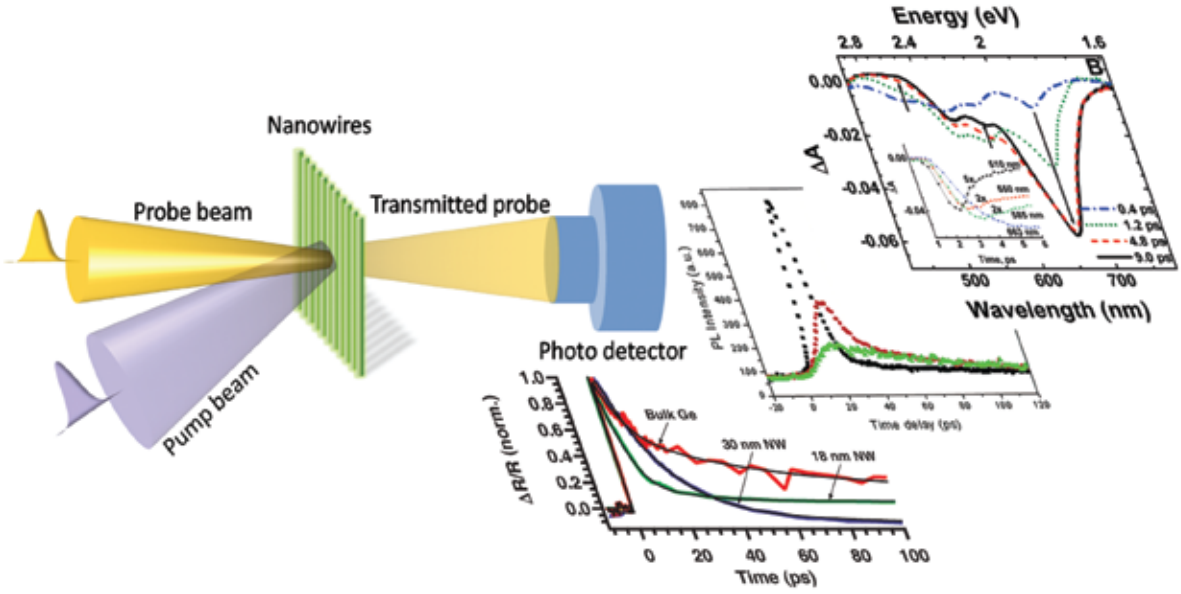
With that drive and enthusiasm, expect to see a finer picture of the nanoscale world from this scientist and photographer.

Ultrafast carrier dynamics in semiconductor nanowires

Accomplishment: Research in nanoscience and nanotechnology is driven by the technological imperative to reduce electronic and photonic devices to smaller dimensions. Semiconductor nanowires have therefore attracted increasing levels of attention, creating an urgent need for sophisticated experimental and theoretical techniques capable of unraveling complex processes in these nanosystems. In the past decade several bottom-up synthesis methods have been developed, allowing for the controlled synthesis of quasi-one-dimensional (1D) nanostructures over a wide range of parameters. The quasi-1D geometry is particularly appropriate for applications such as nanowire lasing and waveguiding, while the size scale of these nanostructures makes them promising candidates for sub-wavelength photonics integration.

In a review paper featured as the cover article of Physica Status Solidi B, Prasankumar, Upadhya, and Taylor explain how ultrafast optical spectroscopy can be used to temporally resolve carrier dynamics in semiconductor nanowires on a femtosecond time scale. Carriers can thus be tracked through their complete life cycle, from their creation by ultrashort pulsed photoexcitation to their eventual destruction through radiative and/or non-radiative pathways. This capability allows researchers to examine the influence of intrinsic and extrinsic parameters, particularly quasi-1D carrier confinement, on these processes with a view toward uncovering new phenomena in these nanosystems. The knowledge gained from these studies could potentially enable the design of nanowires and/or their heterostructures to achieve a desired functionality, harnessing the vast potential of nanotechnology to make an impact in real world applications.

Significance: Ultrafast optical spectroscopy is a powerful tool for investigating nanostructures, as it can be used to examine different material properties and unravel the links between them in the time domain. This has been particularly useful when investigating carrier dynamics in semiconductor nanowires, a major focus of our research at CINT that cuts across several thrusts, as evidenced through the nanowire IFA. We have shown that ultrafast optical measurements can be used to track carrier dynamics in semiconductor nanowires and extract device-relevant parameters such as the diffusion length and surface recombination velocity in these systems, which will be important for future applications in areas such as photovoltaics, thermoelectrics, and nanophotonics.



A schematic of an ultrafast optical pump-probe measurement on an ensemble of semiconductor nanowires. The figures shown on the right demonstrate the ability of ultrafast optical spectroscopy to track the temporal evolution of carrier populations in semiconductor nanowires with femtosecond time resolution as a function of intrinsic parameters (e.g., the nanowire diameter) and extrinsic parameters (e.g., the wavelength or excitation fluence), providing vital information that cannot be obtained by any other technique.

Attribution: User Project Number C2009A087

Publications:

R. P. Prasankumar, P. C. Upadhya, and A. J. Taylor, "Ultrafast dynamics in semiconductor nanowires," invited review article, Physica Status Solidi (b), 246, 1973 (2009).

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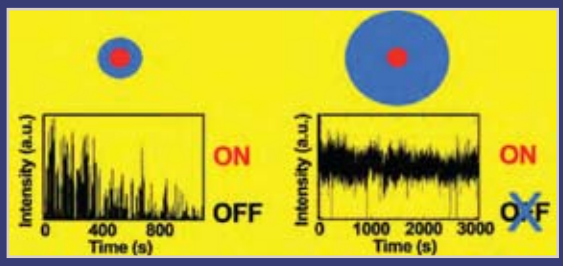
"Giant" Nanocrystal Quantum Dots:

Suppressed Blinking and Auger Recombination through Solution-Phase Physical and Electronic-Structure Engineering

Accomplishment: In many respects, semiconductor nanocrystal quantum dots (NQDs) are near-ideal "building blocks" for light-emission applications. Their optically excited emission is efficient (quantum efficiencies can approach unity), narrow-band ("color-pure"), and particle-size-tunable, i.e., light-output colors are precisely tunable from the ultraviolet through the visible and into the mid-infrared depending on NQD composition and size. Furthermore, NQDs are synthesized using scalable and inexpensive solution-phase approaches, rendering NQDs with "molecule-like" properties and an ability to be chemically processed. Despite these enabling characteristics, conventional NQD optical properties are sensitive to NQD surface chemistry and chemical environment, which has three important results: (1) high NQD solution-phase photoluminescence quantum yields are not maintained in the solid state (e.g., 90% solution-phase QYs can drop to ~10% when the NQDs are deposited into thin-film form), (2) NQDs "photobleach" (PL intensity degrades over time), and (3) NQDs at the single-particle level "blink" (exhibit fluorescence intermittency), all limiting total NQD "brightness."

We recently reported that these key NQD optical properties can be rendered independent of NQD surface chemistry and chemical environment by growth of a very thick, defect-free inorganic shell.1 Effectively, we isolated the wavefunction of the NQD core from its surface, creating a colloidal NQD that is structurally more akin to physically grown epitaxial QDs. We named this new functional "class" of NQD the "giant" NQD (g-NQD). Importantly, g-NQDs are insensitive to changes in surface chemistry, do not photobleach, and show markedly improved blinking behavior (Figure) – a long sought-after goal for this important group of light emitters. To date, up to 50% of a given sample is "non-blinking," where non-blinking is defined as the NQD being "on" under continuous excitation for >99% of the long observation time of 54 minutes.2,3 As follow-on to these initial studies, we performed ensemble photoluminescence dynamical studies that revealed direct evidence for strong suppression of non-radiative Auger recombination (including biexciton lifetimes that are 50 times longer than those obtained for conventional NQDs) and an indication that subtle structural/electronic features of our g-NQDs were likely causing the exceptional photophysics.4 Lastly, we have also observed unprecedented emission from multiexciton states in low-temperature single-g-NQD photoluminescence studies. In the case of conventional NQDs, multiexciton states cannot participate in emission, while for g-NQDs, multiexciton-state emission is in fact efficient.

Significance: These latest studies performed at CINT demonstrate that g-NQDs can afford new exciton→photon conversion pathways. In this way, g-NQDs comprise ideal "test subjects" for NPON Thrust studies of light/matter interactions and energy-conversion processes. In addition, the "robustness" of g-NQDs make them ideal building blocks for integration into micro/macroscale device architectures.



(a). Fluorescence time trace for a representative conventional commercial CdSe core/shell NQD (left) and a CdSe/CdS g-NQD (right) with an experimental temporal resolution of 200 ms.

Attribution: Work derived in part from User Project U2008A109.

Publications

Chen, Y., Vela, J., Htoon, H., Casson, J. L., Werder, D. J., Bussian, D. A., Klimov, V. I., and Hollingsworth, J. A.,""Giant" multishell CdSe nanocrystal quantum dots with suppressed blinking," J. Am. Chem. Soc. 130, 5026 (2008).

Hollingsworth, J. A., Vela, J., Chen, Y., Htoon, H., Klimov, V. I., and Casson, A. R., "Giant multishell CdSe nanocrystal quantum dots with suppressed blinking: novel fluorescent probes for real-time detection of single-molecule events," Proc. SPIE 7189, 718904 (2009).

Hollingsworth, J. A.; Chen, Y.; Vela, J.; Htoon, H.; Klimov, V. U.S. Patent Application No. 61/065,077 (Feb. 2009): "Thick-shell Nanocrystal Quantum Dots."

García-Santamaría, F.; Chen, Y.; Vela, J.; Schaller, R. D.; Hollingsworth, J. A., and Klimov, V. I "Suppressed Auger Recombination in "Giant" Nanocrystals Boosts Optical Gain Performance," Nano Lett., 9, 3482 (2009).

External coverage: Nature Research Highlight: Nature 452, 916 (2008).

Soft, Biological and Composite Nanomaterials

The Soft, Biological and Composite Nanomaterials (SBCN) thrust focuses facilities and expertise on solution-based, "bottom-up" approaches for development of integrated nanomaterials. Synthesis, assembly, and characterization of soft or biological components and the integration of these components across multiple length scales to form functional architectures are of interest. High-level topics include the intersection of materials science with biology, the interfacial science of soft and composite materials, active- and self-assembly methods, soft/hard/bio composite materials, systems integration, and advanced characterization techniques.

Ongoing research consists of development of molecular and biomolecular recognition methods for materials assembly, development of transduction strategies for molecular-scale events, chemical and biomolecular functionalization of interfaces to control assembly and interactions of components, passive and active assembly of nanomaterials with complex and emergent behavior, single-particle and single-molecule spectroscopy, interfacial characterization, imaging, and microfluidic platform development.

Capabilities and expertise within the thrust include molecular biology, organic and inorganic synthetic chemistry, surface chemistry, spectroscopy, single-molecule detection, scanning probe microscopy, optical imaging, and the design of microfluidic systems and other integrated architectures (including CINT Discovery Platforms™). Major facilities include laboratories and instrumentation for biochemistry, cell-culturing and biomolecular engineering, self-assembled material and thin-film preparation, chemical synthesis, scanning probe microscopies, optical microscopy and spectroscopy, Langmuir-Blodgett troughs, interfacial force measurements, and spectroscopic or imaging ellipsometry.

Scientific directions include:

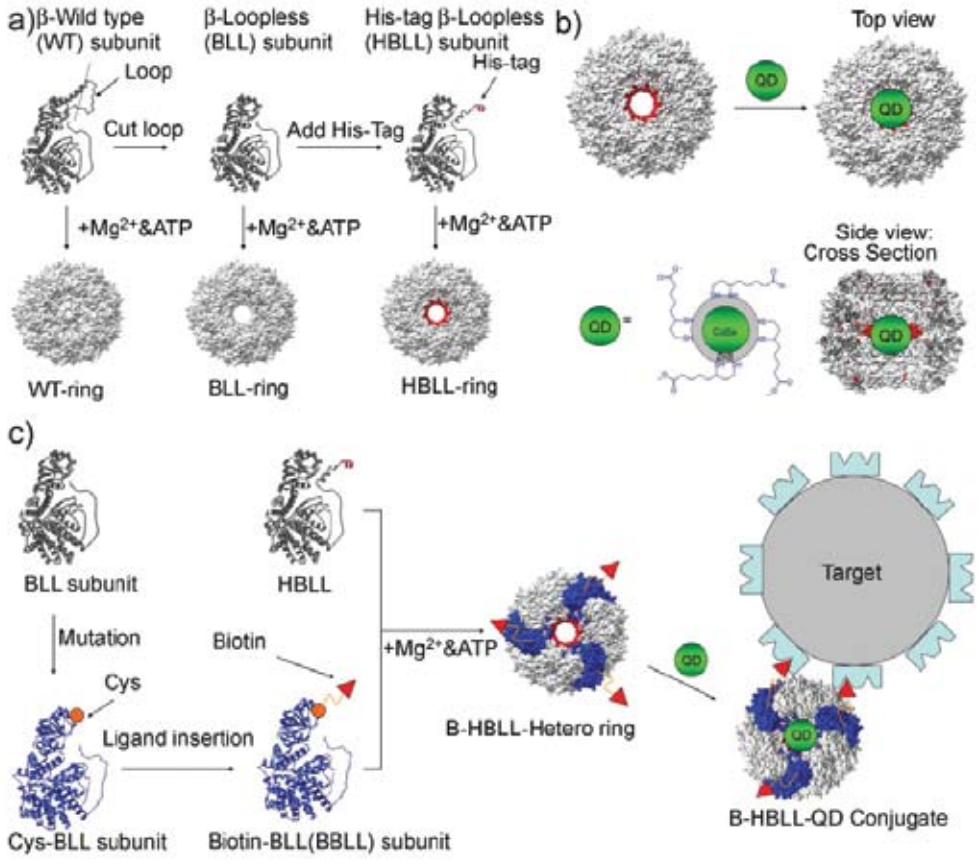
- Molecular and biomolecular recognition
- Surface functionalization involving monolayers, lipids, and membranes
- Active, passive, and hierarchical assembly of solution-based nanocomposites
- Emergent behavior in bio-inspired materials
- Integration of active biomaterials and nanocomposites into microfluidic systems

An intrinsically fluorescent recognition ligand scaffold based on chaperonin protein and semiconductor quantum-dot conjugates

Accomplishment: The self-assembly of materials into ordered structures is a grand challenge for bottom-up fabrication. Likewise, the functionalization of materials with biological molecules is an important step in production of mixed composites for self-assembly, or for biological or chemical sensing. Biological molecules (i.e. DNA or antibodies), by virtue of their programmed molecular recognition, may be ideal molecules for the functionalization of nanomaterials and for the assembly of materials into higher ordered structures. Toward this broader goal of CINT, CINT-users Cotlet, Swanson, Trent, and Xie have genetically modified a chaperonin protein from the hyperthermophilic Achaean Sulfolobus shibata. This chaperonin is a quaternary assembly of proteins with dimensions of 17 nm x 16 nm, and with a central cavity. Here the investigators mutated individual proteins of the assembly to do three things: 1) enlarge the cavity of the protein assembly to accommodate a quantum dot of defined size; 2) incorporate a His-tag within the cavity for the coordination of the inorganic quantum dot; and 3) incorporate a chemical moiety on the surface of one protein to functionalize the nanobio assembly for sensing.

These investigators found that the wild-type chaperonin, with a small internal cavity, could not accommodate a small quantum dot (QD520 nm); whereas, the modified assembly, with a 9 nm cavity, could accommodate a small quantum dot. Xie et al, have shown that the quantum dot-chaperonin assembly has exceptional dissociation constants and attribute the stability of the complex and association energy to the ligand exchange of the His-tag on the chaperonin for the DHLA ligands on the quantum dot, thereby producing a strong conjugate. Interestingly, once protected in the internal cavity, the quantum dot showed not only increased stability from aggregation, but more importantly greatly enhanced photoluminescence. From detailed photophysical characterization the authors were able to show that the increased fluorescence results from a decrease in the nonradiative processes (i.e. blinking) of the quantum dot.

Significance: The authors have created a protein that can self-assemble and incorporate a nanomaterial (quantum dot). As these chaperonin proteins are known to form 2D arrays, this work builds toward producing patterned nanoparticles. Further, the external functionalization could aid in directed patterning or for the production of a sensor.



Attribution: Work derived in part from user project: U2008A167

Publication:

"An intrinsically fluorescent recognition ligand scaffold based on chaperonin protein and semiconductor quantum-dot conjugates" H. Xie, Y.-F. Li, H.K. Kagawa, J.D. Trent, K. Mudalige, M. Cotlet, and B.I. Swanson Small, 2009, 5, 1036-1042.

CINT Scientist Highlight - Jennifer Martinez

At home on her family's land in southern Colorado, Jennifer Martinez fixes fences and digs ditches. At home in her laboratory at the Center for Integrated Nanotechnologies (CINT) at Los Alamos National Laboratory, Martinez designs and characterizes the interface between inorganic and biological systems using biomolecular recognition strategies, with the goal of creating functional nanoscale materials. A dichotomy perhaps, but not when you consider she grew up in a family of outdoorsmen. "There's not a scientist in my family, but my dad—and my whole family—are good naturalists," Martinez said. "Perhaps that's where I get my inspiration."

For this as well as other inspired research, Martinez was recently recognized with a prestigious Presidential Early Career Award for Scientists and Engineers. The award is the highest honor bestowed by the U.S. government to outstanding scientists early in their careers. She was one of eight researchers funded by the Department of Energy's Office of Science and the National Nuclear Security Administration to be recognized, and one of 68 researchers supported by nine federal departments and agencies to receive the award. Martinez said she appreciates the honor, "although I can think of many others here at Los Alamos who are as—or more—deserving," and considers it "a great incentive to do some nice work."

Martinez, a staff scientist in CINT, received a bachelor of science degree in chemistry from the University of Utah and a doctoral degree in bioinorganic chemistry from the University of California, Santa Barbara. She joined the Laboratory in 2002 as a Director's Postdoctoral Fellow, performing biological sensing research, which has applications in medical diagnostics as well as in the detection of biological threat agents. She described the work as "an incredible challenge" that allowed her to work with a diverse set of people. "Los Alamos has really great scientists…who are used to collaborating with each other," she said. "People are engaged. They are likely to talk to you about the science and want to get involved in new science."

Martinez finds that spirit of cooperation energizing, and at CINT, she said, a walk down the hall and a knock on a door puts her in touch with people from a range of disciplines—from theorists to biochemists to spectroscopists. "To have so many people with very diverse backgrounds in one building, working in one group, is amazing."

Martinez now splits her time between working with visiting scientists who come to CINT to perform cutting-edge nanoscience and with her Los Alamos colleagues on a range of basic research projects. "Jennifer's very valuable in terms of bringing in new users from the external scientific community and ensuring they have a good experience while working here at CINT, and being able to move ahead projects that are quite interdisciplinary in nature," said Andrew Shreve, a leader in the soft, biological and composite nanomaterials area of CINT. Shreve works together with Martinez on a project using biological strategies to develop new materials.

Martinez's research on developing high-yield synthesis of metal nanoclusters, in which she collaborates with Shreve, Brian Dyer, Jim Werner, Dung Vu, Sergei Ivanov, and Andrei Piryatinski, has biosensing applications and could eventually lead to better cancer detection techniques. Her research into biocompatible polymers, in which she works with Emily Schmidt, Andrew Bradbury, and Csaba Kiss, could lead to stronger, flexible, highly biocompatible materials of interest to the medical community.

Her work has been published in high-profile journals such as Science, Proceedings of the National Academy of Sciences, Journal of the American Chemical Society, and Langmuir. "Jennifer is one of those people who adds to our capability base tremendously," said Basil Swanson, one of Martinez's former postdoctoral mentors and a Chemistry Division staff member. Much of the research at Los Alamos requires a cross-discipline effort, Swanson said, and "she is able to talk the language of the people— the engineers, who help us develop our (devices and products) and the people in the hard-core sciences."

Spreading of Liquid Polymer Droplets on Liquid Surfaces

Accomplishment: While the spreading of droplets on solid substrates has been intensively explored much less is known regarding the spreading process on imbueable interfaces. On a liquid substrate, the liquid simultaneously spreads and permeates the interface, a process that takes place at a large variety of interfaces from biomembranes to thin coatings. Our simulations1 for liquid oligomer films on a homopolymer film have shown that the degree of interpenetration and amount of damping from the film depends strongly on the viscosity of the film and the relative interaction of the two liquids. For very viscous films a precursor foot spreads ahead of the film as shown in right panel of Figure 1. For less viscous films the drop penetrates the film while spreading and there is no precursor foot (left panel Figure 1).

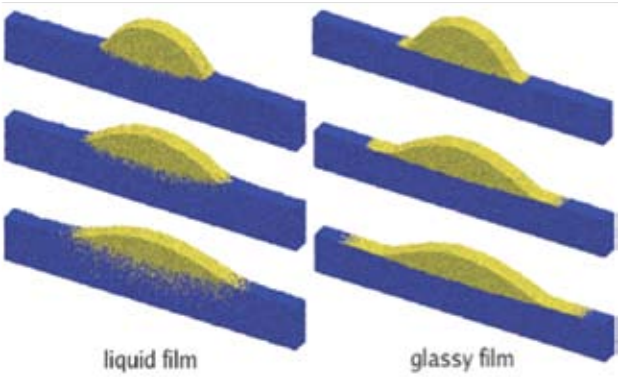


Figure 1. Liquid polymer droplet (yellow) spreading on liquid (left-blue) and glassy film (right-blue) at t = 4000, 16000 and 32000τ (top to bottom), where τ is unit of time in Lennard-Jones units.1

To distinguish the relative roles of the spreading from the interdiffusion we also studied the interpenetration of two polymer films. Our initial study examined the interdiffusion of short oligomers into entangled polymeric matrices.2 As the chain length of the oligomer increased from that of a simple liquid to an unentangled polymer there was a distinct change in the shape of the polymer and oligomer profiles due to the swelling of the polymer film. When both films are composed of long entangled polymer chains, we find an extended, early time regime in which the mass uptake scales as ta with a ~0.19.3 Unlike the dynamics in a polymer melt which is dominated by the reptation motion of the monomers at intermediate times, interdiffusion is dominated by motion of the chain ends. A primitive path analysis of the chains showed for the first time the development of entanglements between the two films as the polymer chains interdiffuse (Figure 2).

Significance: Control over spreading on responsive surfaces such as polymers and biomembranes is highly desirable for a large variety of current and emerging applications from responsive coating to drug delivery. The focus of the present work has been to identify the various dissipation mechanics for spreading droplets on liquid surfaces. For two entangled polymer film, this work gives new insight into the technologically important process of welding and self-healing of polymer films.

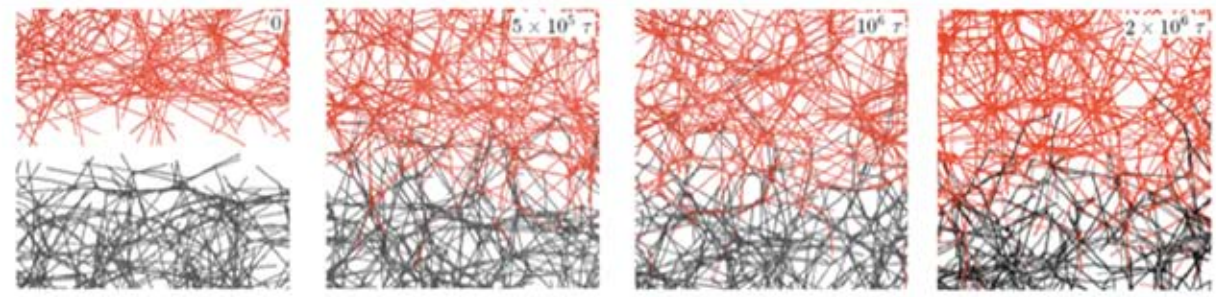


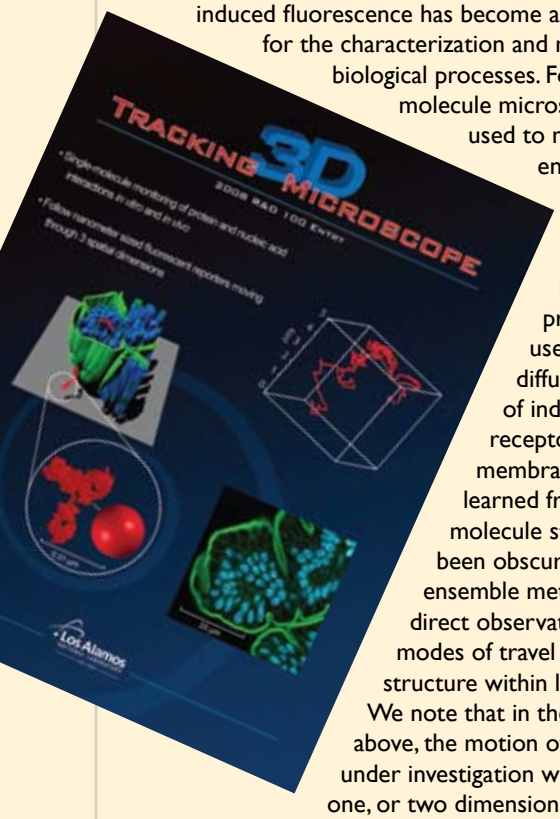
Figure 2. Snapshots of thin chain primitive path mesh for interpenetration of two films each of chain length N = 500 beads.3 Chains in upper film are colored red, chains in lower film black.

Attribution: User Projects C2008A139 and C2009A066.

Publications:

Spreading of Liquid Polymer Droplets on a Permeable Polymer Liquid, F. Pierce, D. Perahia, and G. S. Grest, EPL 86, 64004 (2009).
Interdiffusion of Short Chain Oligomers into an Entangled Polymer Film, F. Pierce, D. Perahia, and G. S. Grest, Macromolecules 42, 3186 (2009).
Self-Healing of Entangled Liquid Polymer Melts, F. Pierce, D. Perahia, and G. S. Grest, submitted for publication (2010).

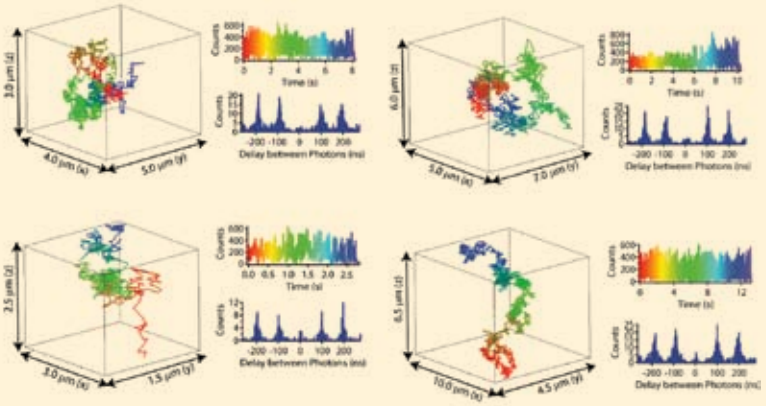
Three Dimensional Tracking of Individual Quantum dots



Accomplishment: The detection of single molecules by laser-induced fluorescence has become a powerful tool for the characterization and measurement of biological processes. For example, single molecule microscopy has been used to monitor individual enzymatic turnovers, directly observe the hand over hand motion of individual motor proteins, and has been used to visualize the diffusion and transport of individual lipids and receptors on live cell membranes. Much has been learned from these single-molecule studies that had been obscured in conventional ensemble methods, such as the direct observation of unexpected modes of travel around domain structure within live cell membranes. We note that in the examples cited above, the motion of the molecule under investigation was limited to zero, one, or two dimensions. We point out perhaps the obvious: most aspects of life, including intracellular signaling and trafficking, are inherently 3-dimensional. However, tracking a single fluorescent molecule or a single quantum dot traveling through 3 dimensional space is a difficult (and until quite recently), an unsolved technical problem.

In 2007, our group, and two other leading spectroscopy labs, independently published methods using confocal feedback that demonstrated 3D tracking of individual quantum dots moving at biologically relevant transport rates. Since our initial experimental demonstration of 3D tracking single quantum dots, we have verified our tracking methods can be done in high background environments and live cells. In 2008, our 3D tracking methods earned an R&D 100 Award. In 2009, our instrumentation was awarded a US Patent

Significance: The development of tools to explore nano-scale spatial heterogeneity in complex, dynamic systems are an important aspect of nanoscience integration. In particular, our 3D tracking methods can be used for characterization of new nanomaterials, can lead to a better understanding of self assembly processes important for a broad range of functional soft nanomaterials, and can directly observe dynamic re-arrangement in planar lipid bilayer architectures (an Integration Focal Area).



Photon pair correlations (anti-bunching) obtained while tracking

Attribution: This technology grew out of internal CINT science, but is currently being used by a number of members of the external user community, including Diane Lidke at the University of New Mexico (U2008A062) and Christine Payne from Georgia Tech (U2008B023).

Publications and Patents:

Lessard, G., P.M. Goodwin, and J.H. Werner, Three dimensional tracking of individual quantum dots Applied Physics Letters, 2007. 91(22): p. 2224106.

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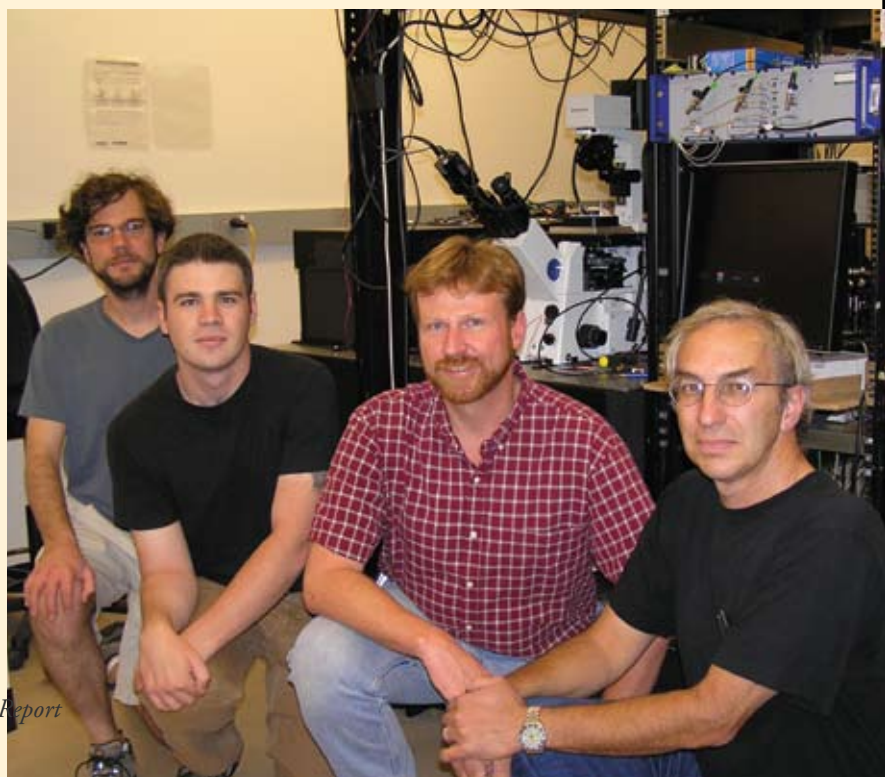
Cang, H., C. Xu, D. Montiel, and H. Yang, Guiding a confocal microscope by single fluorescent nanoparticles. Optics Letters, 2007. 32(18): p. 2729-2731.

Wells, N.P., G.A. Lessard, and J.H. Werner, Confocal, 3-Dimensional Tracking of Individual Quantum-Dots in High Background Environments. Analytical Chemistry, 2008. 80: p. 9830-9834.

Wells, N.P., G.A. Lessard, M.E. Phipps, P.M. Goodwin, D.S. Lidke, B.S. Wilson, and J.H. Werner, Going Beyond 2D: Following membrane diffusion and topography in the IgE–Fc[Epsilon]RI system using 3-dimensional tracking microscopy. Proc. of the SPIE, 2009. 7185: p. 7185-1 to 7185-13.

Werner, J.H., P.M. Goodwin, and G.A. Lessard, Apparatus and method for tracking a molecule or particle in three dimensions, in US Patent 7,498,551. 2009, Los Alamos National Laboratory: USA.

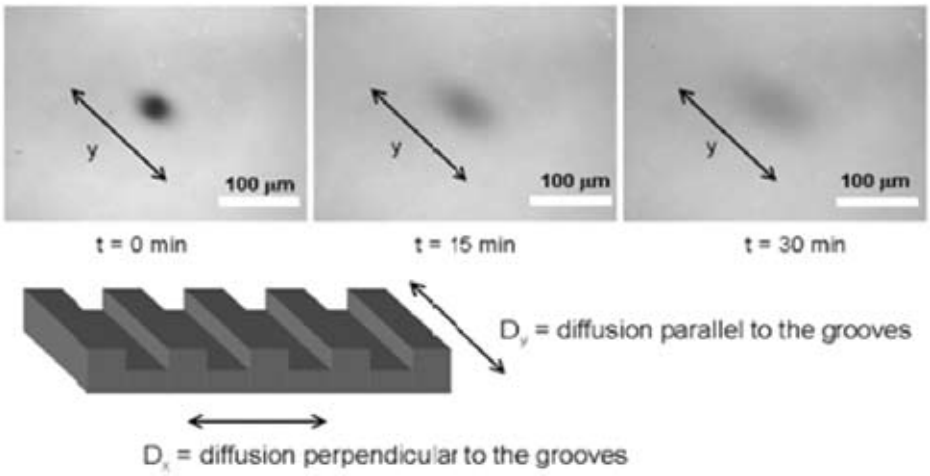


Formation and Dynamics of Supported Phospholipid Membranes on a Periodic Nanotextured Substrate

Accomplishment: Supported planar lipid bilayers are important model systems enabling the control, organization, and study of membranes and membrane-associated proteins. In addition to fundamental biophysical investigations, supported bilayers are useful scaffolds for many practical applications. For many of these applications, there is often a need to physically segregate and isolate sections of planar lipid bilayers on nanometer tomicrometer length scales. Patterning methods have successfully been used to synthesize two-dimensional lipid bilayer constructs and have been well characterized, however efforts to synthesize and characterize three-dimensional lipid bilayer constructs have been limited. In this work, we used nanotextured architectures of ordered periodicity to investigate lipid bilayer dynamics in three dimensions in order to better determine use in biophysical applications and potential membrane device applications.

We used a combination of AFM and quantitative fluorescence microscopy to demonstrate the formation of POPC bilayers conformed to a nanotextured surface. Figure 1 exhibits the anisotropic recovery of a POPC LBA on the nanotextured surface. One of the more important findings of this work is a demonstration of how quantitative analysis of anisotropic diffusion on the micrometer scale can yield information concerning the underlying nanoscale morphology. In addition, this work lays the foundation for future studies directed at how nanotextured surfaces with tunable physical or chemical properties might be used to influence membrane structure, dynamics, and functions as well as lead to development of platforms for membrane protein incorporation and investigation.

Significance: Lipid membrane architectures have potential in many biophysical studies as well as biosensor and bio-hybrid devices. A focus of our soft, biological and nanocomposite thrust and membrane integrations focus area is to utilize lipid membrane architectures for advanced biophysical studies such as single molecule and three-dimensional tracking of membrane components as well as synthesis of responsive bio-hybrid nanomaterials. The represented work shows a useful platform that is currently being studied for both of these research efforts.



Fluorescence recovery after photobleaching showing anisotropic recovery of POPC lipid bilayer on nanotextured architectures.

Attribution: User Project Number U2008A083
Publication:
J. Werner, G.A. Montaño, A Zurek, A. Garcia, E. Akhadov, G.L Lopez, A.P. Shreve. (2009) Formation and characterization of a supported phospholipid membrane on a periodic nanotextured substrate. Langmuir 25(5):2986-2993.

Theory and Simulation of Nanoscale Phenomena

The Theory and Simulation of Nanoscale Phenomena (TSNP) thrust focuses on the theoretical and simulation understanding of the fundamental nanoscale phenomena that underlie integrated nanomaterials. Classical and quantum methods are applied to determine the emergent properties of nanoscale materials and systems as a basis of assembly of nanomaterials.

A key area of interest is the interfacial interactions that are unique to the nanoscale and are significant in determining the material properties. Material properties which are presently being studied include energy harvesting and transport, molecular electronics, mechanical properties of biopolymer networks, and dynamical transport. Ongoing research topics include passive and active assembly of nanomaterials with complex and emergent behavior, theoretical spectroscopy and nonlinear optical response, energy transfer and charge transport, DNA nanoelectronics, local defects, optical and tunneling probes, mechanical properties of nanocomposites.

Capabilities and expertise within the thrust include molecular theory methods, atomistic and coarse-grained molecular dynamics simulations, static and time dependent density functional theory, many body quantum methods. Major facilities include visualization hardware and software and parallel computers.

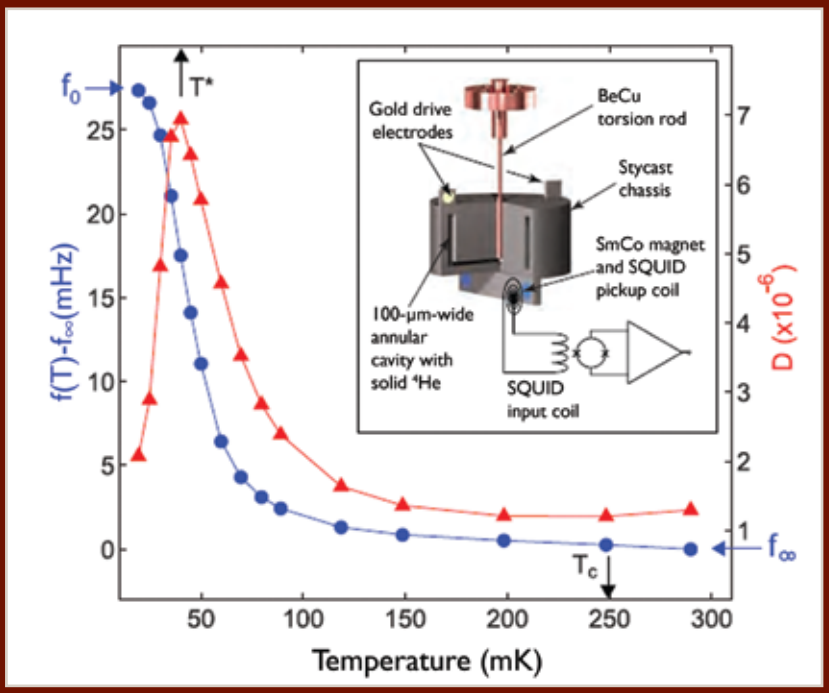
Scientific directions include:

Assembly of nanomaterials; interfacial interactions
Emergent properties of nanoscale materials and systems
Electronic, magnetic, and optical properties at the nanoscale

Cracking the Supersolid

Accomplishment: Recent experimental and theoretical work shows that frozen helium must have atomic disorder rather than perfect crystallinity for it to become a supersolid. The discovery of the putative supersolid state in the bosonic solid 4He in 2004 has been compared to the Nobel-Prize-winning discovery of superfluidity in fermionic 3He in 1972. Kim and Chan observed an anomaly in the mechanical torsional oscillator subjected to a driving torque. The anomaly in and of itself is very small and sets in at quite low temperatures, roughly at 200 mK above absolute zero temperature and at pressures above 25 bar. This anomaly persists upon applying pressure to the cell up to 160 bar. To put this excitement in perspective: For the last 40 years an intensive search for a supersolid state was unsuccessful. The observation by Kim and Chan started a renaissance of new measurements and theories in this field to prove the existence of this exotic state of matter. The supersolid phase is one of the enigmatic quantum states of matter, whereby the very same atoms exhibit simultaneously crystalline order, with long-range order in the density modulation and at the same time off-diagonal long-range order, best characterized as some kind of superfluid stiffness that would allow atoms to flow without resistance in response to smooth changes of the superfluid phase. The richness of quantum mechanics in this quantum solid allows the very same atoms to be partially localized, i.e., locked in the lattice, and yet to be delocalized and partially flow in response to small pressure gradients.

Significance: In these Science articles by Phillips and Balatsky and by Hunt et al the role of defects, long relaxation times and interactions between defects in formation of a possible superglass state in solid He4 at lowest temperatures are addressed.



The resonant frequency shift f (T) − ϕ∞ (blue circles) and dissipation D(T) ≡ Q−1 (T) (red triangles) for our TO-solid 4He system. Indicated with black arrows are T, the temperature at which D(T) peaks and the slope of f (T) − ϕ∞ is maximal, and Tc, the temperature at which a change in f (T) − ϕ∞ becomes detectable above the noise. (Inset) A schematic of the superconducting quantum interference device (SQUID)–based torsional oscillator (TO). Applying an ac voltage to the drive electrodes rotates the Stycast chassis (containing the solid 4He in a 100-mmwide annular cavity of radius 4.5 mm) about the axis of the BeCu torsion rod. The angular displacement of a SmCo magnet mounted on the TO generates a change in the magnetic flux through the stationary pickup and input coils of a dc-SQUID circuit and thereby a voltage proportional to displacement.*

Attribution: Work derived in part from User Project U2008B063

Publications

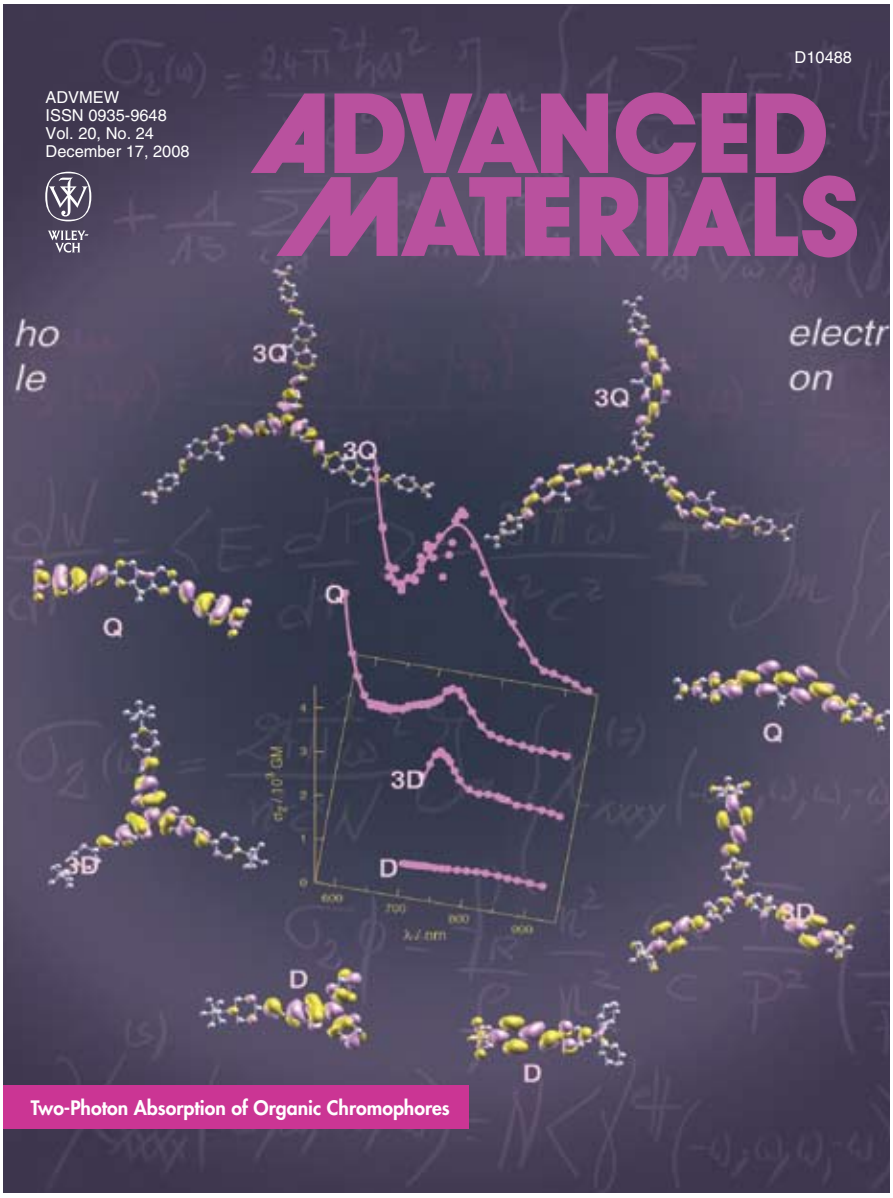
P. Phillips and A. V. Balatsky. Cracking the Supersolid. Science 2007, 316, 1435.

B. Hunt, E. Pratt, V. Gadagkar, M. Yamashita, A. V. Balatsky, and J. C. Davis. Evidence for a Superglass State in He-4. Science 2009, 324, 632.

Enhanced Two-Photon Absorption of Organic Chromophores:

Theoretical and Experimental Assessments

Accomplishment: We review experimental and theoretical methodologies allowing detailed investigation and analysis of two-photon absorption (TPA) properties of organic chromophores. This includes femtosecond two-photon excited fluorescence experimental setups and quantum-chemical methodologies based on time-dependent density functional theory. We overview of theoretical approaches used for modeling of TPA properties in organic chromophores including first principle quantum-chemical methods and effective reduced models such as Frenkel exciton and a few states models and analyze of range of applicability of these techniques. In addition, experimental methodologies including including Z-scan, nano-, pico-, and femto-second TPEF are reviewed with special attention to sources of errors that yield improper evaluation of TPA cross sections. Finally, we thoroughly analyze physical phenomena and trends leading to large two-photon absorption responses of a few series of model chromophores focusing on the effects of symmetric and asymmetric donor/acceptor substitution and branching.



Significance: Over the past decade, there has been a broad research activity focused on organic materials with enhanced two-photon absorption (TPA) properties. Such materials lead to new technologies in the fields of chemistry, biology, and photonics, such as 3-dimensional printing, non-destructive bio-imaging and photodynamics therapy. To make possible these applications, an enormous body of knowledge stemming from experimental and theoretical studies has been accumulated. Here we summarize the research quest for efficient organic TPA chomophores.

Two-photon absorption offers a rich playground for development of novel optical multifunctional technologies. The cover page of the journal shows experimental TPA spectra of several molecules complemented by theoretical analysis of participating excited states. This allows formulating specific structure-property relationships to achieve desirable nonlinear optical response of organic molecules.

Attribution: User Project Number C2008A031

Publication:

F. Terenziani, C. Katan, M. Blanchard-Desce, E. Badaeva, and S. Tretiak, "Enhanced two-photon absorption of organic chromophores: theoretical and experimental assessments", Advanced Materials 20 1-38 (2008).

Simulations of nanotribology with realistic probe tip models

Accomplishment: Using massively parallel molecular dynamics simulations, we are able to understand the nanotribological properties of alkylsilane self-assembled monolayers (SAMs) on amorphous silica. In contrast to studies with opposing flat plates, as found in the bulk of the simulation literature, we use a model system with a realistic AFM tip (radius of curvature ranging from 3 to 30 nm) in contact with a SAM-coated silica substrate. We compare the differences in response between systems in which chains are fully physisorbed, fully chemisorbed, and systems with a mixture of the two. Our results demonstrate that the ubiquitous JKR and DMT models do not accurately describe the contact mechanics of these systems. In shear simulations, we find that the chain length has minimal effects on both the friction force and coefficient. The tip radius affects the friction force only (i.e., the coefficient is unchanged) by a constant shift in magnitude due to the increase in pull-off force with increasing radius. We also find that at extremely low loads, on the order of 10 nN, shearing from the tip causes damage to the physisorbed monolayers by removal of molecules.

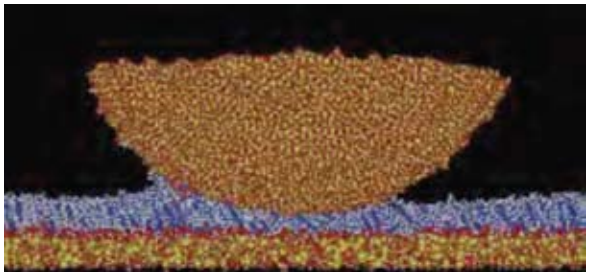


Figure 1: Snapshot for alkylsilane self-assembled monolayer (blue and white) in contact with a tip that has a 10 nm radius after 2 ns of shear at a velocity of 2 m/s and normal load of 10 nN. A 4 nm slice from the center of the SAM is shown with all atoms in the tip.

Attribution: CINT science

Publication:

M. Chandross, C.D. Lorenz, Mark J. Stevens and G.S. Grest, "Simulations of Nanotribology with Realistic Probe Tip Models," Langmuir 24, 1240 (2008).

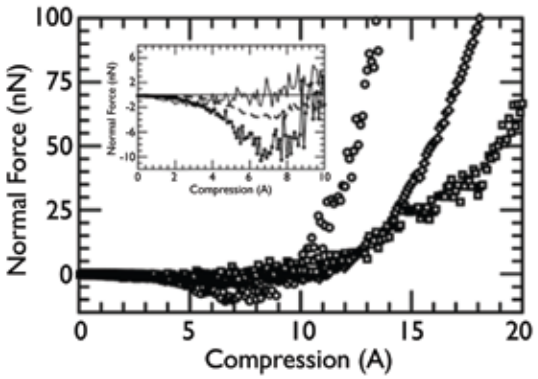


Figure 2: Normal force as a function of compression for a tip in contact with alkylsilane SAM for the □ 3 nm, ◇ 10 nm and ○ 30 nm tip, respectively. The inset shows an enlargement of the attractive regime with the 3 nm, 10 nm and 30 nm tips shown as solid line, dashed line and solid line with points, respectively (lines here simply connect points to guide the eye). The zero of the x-axis is a tip-SAM separation of approximately 1.5 nm. The compression rate is 5 m/s.

Significance: Tip-based nanotribology simulations bring two major changes as compared to flat-plate simulations, namely load-dependant contact areas, and the possibility of the removal of material from the substrate. In the case of material removal, our simulations demonstrate that, at least for fully physisorbed monolayers, even extremely low loads can lead to damage to the monolayer. In addition, this work paves the way to treat nanoparticle interactions using all atom simulations. By changing the tip from a hemisphere to a full sphere, the tip becomes a nanoparticle interacting with a surface. Since we can do the tip simulation, we can to the nanoparticle simulation.

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S.Thomas Picraux, Laboratory Fellow: Los Alamos National Laboratory, 2009

Mark Stevens, Fellow, American Physical Society, 2009

Antoinette. Taylor, Laboratory Fellow: Los Alamos National Laboratory, 2009

James Werner, Best Paper Award, Single Molecule Session of Photonics West. This was received for "Going beyond 2D: Following membrane diffusion and topography in the IgE–Fc[epsilon]RI system using 3- dimensional tracking microscopy," Proceedings of the SPIE v.7185 2009 7185-1 to 7185-13. Wells, N.P., Lessard G.A., Phipps M.E., Goodwin P.M., Lidke D.S., Wilson B.S., and Werner J.H., 2009.

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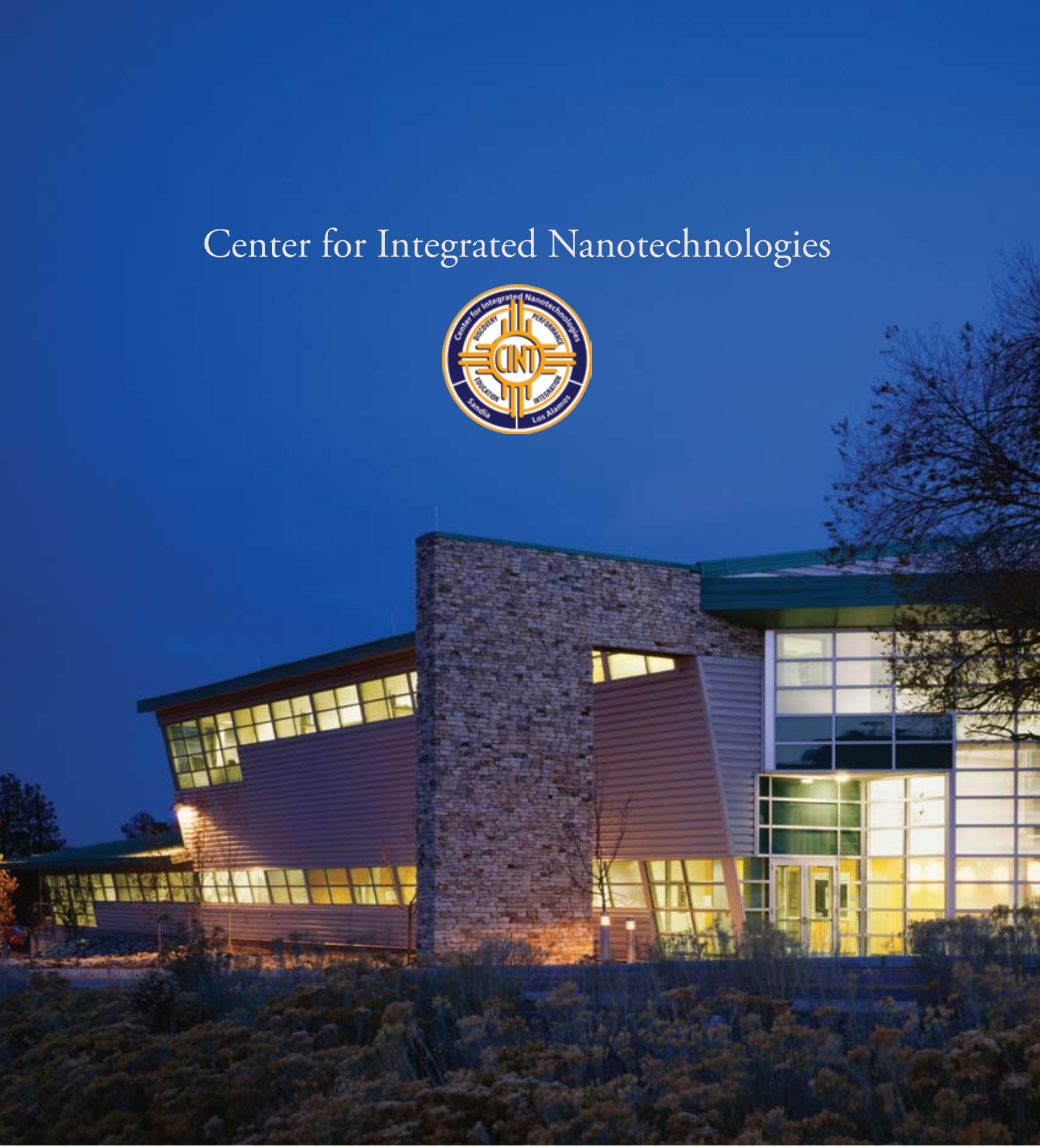

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